ISOTOPES AND RADIATION TECHNOLOGY

A Quarterly Technical Progress Review

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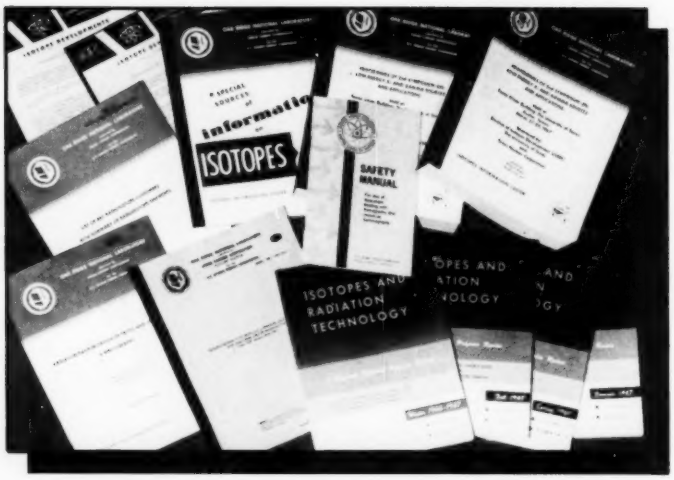

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ISOTOPES INFORMATION CENTER

Isotopes and Radiation Technology

Vol. 6, No. 2 Winter 1968–1969

Contents

I Isotope Production and Development

131 **Radioisotopes: Production and Development of Large-Scale Uses**
131 Summary
138 Production
144 **American Nuclear Corporation Emphasizes Irradiators**
144 Facilities
145 Commercial Products

II Isotope Technology Development

146 **Removal of Radioactive Strontium and Cesium from Certain Vegetables and Fruits During Normal Preparation for Preservation**
146 Methodology
147 Results
149 Discussion
149 **Neutron Radiography Using Nonreactor Sources**
149 Small-Source Thermal-Neutron Radiography: Early Work
150 Fast-Neutron Radiography
150 Thermal-Neutron Radiography: Recent Developments
151 Conclusions

III Food Irradiation

154 **Designing for Lower Food-Irradiation Costs**
155 Source Procurement Needs
156 Comparative Requirements for ^{137}Cs
157 Dose-Distribution Measurements
158 Compact Shellfish Irradiator
160 Overall Cost Estimates
161 Summary
162 **New Food-Irradiation Publications**

IV Process Radiation Development

164 **Study of Radiation Processing**
165 Survey Procedures
165 General Observations
167 Specific Observations
167 Conclusions
169 Bibliography
173 **Extracts from a Report on Grafting of Organic Polymers onto Inorganic Substances**
174 Polymerization and Grafting Mechanisms
177 Adhesion of Polymers to Inorganic Substrates
179 Radiation Polymerization on Inorganic Substances
183 Summary and Conclusions

V Isotopic Heat and Power Development
188 **English Colloquium on Isotopic Thermoelectric Generators**
188 General Principles
189 RIPPLE Applications
192 Properties of Thermocouple Materials
193 Thermoelectric Conversion Theory
195 Civil Aviation Application
195 Marine Navigation Application
196 Other Practical Applications
197 **Isotopic Generators in the Federal Republic of Germany**
197 Thermionic Conversion
198 Thermoelectric Conversion
198 Photoelectric Conversion
199 Thermophotovoltaic Conversion Systems
199 Pacemakers

VI Applications in Life Sciences
200 **Tagging of Small Animals with Radioisotopes for Tracking Purposes**
201 Technique
201 Animals Studied

VII Isotopes Around the World
205 **Recent Isotopic Source Developments in the USSR**
205 Spent-Fuel-Element Source for Vulcanizing Tires
206 Gamma-Irradiation Facility with Cone-Shaped Source
207 "Beta" Series of Isotopic Power Sources
207 State of the Art of Radiation Processing
208 **Practical Applications of Radioisotopes in the USSR**
208 Isotopes in Process Monitoring and Measurement
209 Tagged Atoms
209 Radioisotopes in Agriculture, Biology, and Medicine
210 Radiation Chemistry

VIII Miscellaneous
212 **Availability of Isotopes and Services**
217 **AEC Activities**
219 **General**
222 **Recent Accessions of Isotopes Information Center**

Briefs
210 **Indonesian Radioisotope Application News Bulletin**
210 **Autoradiographic Techniques**
211 **Preparation of a ^{55}Fe Source by Electroplating**
225 **Radioactive Weighing System for Moving Vehicles**
225 **Isotopic Chromatography**

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Isotopes and Radiation Technology

is a Technical Progress Review prepared by the Isotopes Information Center, Oak Ridge National Laboratory, at the request of the Divisions of Technical Information and Isotopes Development, U. S. Atomic Energy Commission. This Review is intended to assist those interested in keeping abreast of significant developments in the fields of isotopes and radiation technology. It is not a comprehensive review of all literature published in this field during a given quarter; rather, it is a mechanism for presenting concise, selected reviews of information on subjects of prevailing Commission interest as it becomes available.

This Review attempts to relate the published results of research and development sponsored by the Division of Isotopes Development to significant developments in radioisotopes and radiation technology, as reported in the world literature. Coverage includes isotope production and development, isotope technology development, isotope applications technology, process radiation development, and radiation processing of food and medical supplies.

In addition to reviews of current literature and special review articles dealing with specific isotopes, facilities, and applications, this publication occasionally contains feature articles prepared by recognized experts on specific topics of current interest. Critical evaluations and interpretations presented are those of the editors and invited reviewers; therefore readers are encouraged to consult the original references in order to obtain all the background of the work reported and the interpretation of the results given by the original authors.

The subject headings listed below have been adopted to help maintain continuity and order, from one issue to the next, in the material reviewed. All reviews, except Feature Articles, will be arranged under these headings; but any one issue will not necessarily contain all the headings.

Isotope Production and Development
Isotope Technology Development
Food Irradiation
Process Radiation Development
Source Development
Isotopic Heat and Power Development
Applications in Life Sciences
Safety
Isotopes Around the World
Miscellaneous

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TECHNICAL PROGRESS REVIEWS

The United States Atomic Energy Commission publishes the Technical Progress Reviews to meet the needs of industry and government for concise summaries of current nuclear developments. Each journal digests and evaluates the latest findings in a specific area of nuclear technology and science. *Nuclear Safety* is a bimonthly journal; the other three are quarterly journals.

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Nuclear Safety
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Reactor and Fuel-Processing Technology
Argonne National Laboratory

Reactor Materials
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Battelle Memorial Institute, Columbus Laboratories

All are available from the U. S. Government Printing Office. See the back cover for ordering instructions.

The views expressed in this publication do not necessarily represent those of the United States Atomic Energy Commission, its divisions or offices, or of any Commission advisory committee or contractor.

Availability of Reports Cited in This Review

United States Atomic Energy Commission (USAEC) reports are available at USAEC depository libraries and are sold by the Clearinghouse for Federal Scientific and Technical Information (CFSTI), National Bureau of Standards, U. S. Department of Commerce, 5285 Port Royal Road, Springfield, Va. 22151. All reports sold by CFSTI are $3.00 for printed copy and $0.65 for microfiche. Each separately bound part of a report is priced as a separate report. Some reports may not be available because of their preliminary nature; however, the information contained in them will generally be found in later progress or topical reports on the subject.

Other U. S. Government agency reports identified in this journal generally are available from CFSTI.

Private-organization reports should be requested from the originator.

United Kingdom Atomic Energy Authority (UKAEA) and Atomic Energy of Canada Limited (AECL) reports are available at USAEC depository libraries. UKAEA reports are sold by Her Majesty's Stationery Office, London; AECL reports are sold by the Scientific Document Distribution Office, Atomic Energy of Canada Limited, Chalk River, Ontario, Canada. UKAEA and AECL reports issued after March 1, 1967, are sold by CFSTI to purchasers in the United States and its territories.

Section

I

Isotopes and Radiation Technology

Isotope Production and Development

Radioisotopes: Production and Development of Large-Scale Uses

Editor's Note: The USAEC has recently published as WASH-1095 a review of nine important radioisotopes, with the title above. Since much of it is appropriate to the scope of *Isotopes and Radiation Technology*, a major portion is being reproduced in two parts, with minor editorial changes. The first part includes an introduction and summary, followed by a discussion of production considerations. The second part, to be published in the next issue [6(3)] of *Isotopes and Radiation Technology*, will cover applications: space power systems; terrestrial power systems; thermal applications; process radiation; and space propulsion.

The Atomic Energy Commission supports a broadly based program to develop a national capability for the large-scale production and use of isotopes as sources of heat and radiation. This program includes numerous applications for peaceful and national defense missions in space; national health programs; conservation of natural resources; and the national program for marine sciences and technology. This article describes the current status, objectives, and program plan for developing these uses of isotopes. The isotopes of main interest in this discussion are described briefly in Table 1.

Large-scale employment of isotopes is just beginning. The anticipated growth in installed capacity of civilian power reactors should provide significant increases in the availability of several important isotopes such as ^{238}Pu, ^{244}Cm, ^{60}Co, and ^{90}Sr.

It is the responsibility of the AEC to encourage use of isotopes as a national resource. Within the AEC effort, research and development required for various applications are coordinated within several programs, each containing projects having common or similar technology requirements.

The body of this article summarizes requirements for production of the various isotopes as part of the background discussion in the section on *Production*. Next, the programs and the interprogram relationships are described. Within each program description the article reviews the competitive factors which influence development objectives and other information necessary for the reader to understand the background of each program being described. Isotopic system characteristics are compared to those of nonisotopic alternatives to illustrate the likely areas of attractiveness and to explain development objectives. Finally, program objectives are presented and the current status of important elements is described.

Summary

Production of Isotopes

Important aspects of the production of isotopes include the forecasting of quantities of isotopes required, the comparison of these quantities with the capacities of existing or projected facilities, and the estimation of production rates and costs which might result from future production programs. Consideration of the special characteristics of each isotope which affect production are of course vital to this planning.

The problems of obtaining reliable forecasts of isotope requirements will be discussed later. Because of the uncertainty to the user about availability and costs of isotopes and the uncertainty to the producer about requirements, the user and producer have to collaborate closely for the most effective joint program planning.

The current limiting factor on the quantity of important radioisotopes that can be produced is the lack of facilities necessary to prepare targets for

ISOTOPES AND RADIATION TECHNOLOGY, Vol. 6, No. 2, Winter 1968–1969

Table 1 Characteristics of Isotopes of Interest

Isotope	Half-life, years	Principal chemical form	Density of (compound), g/cm^3	Principal radiation	MPC,* μc per cm^3	Watts/gram (pure isotope)	Watts/gram (compound)	Power density (compound), watts/cm^3	Curies per watt	Cost range, $/thermal watt†	Shielding required‡	Remarks
^{60}Co	5.3	Co metal	8.7	γ	3×10^{-9}	17.4	1.7	15.2	65	7–25	Heavy (9.5)	Currently in large-scale use principally as radiation source
^{90}Sr	28	$SrTiO_3$	3.7	β,x	3×10^{-10}	0.95	0.22	0.82	148	25–35	Heavy (6)	Reference terrestrial power fuel
^{137}Cs	30	CsCl	3.6	β,γ	5×10^{-9}	0.42	0.12	0.42	207	20–30	Heavy (4.6)	Alternative to ^{60}Co
^{144}Ce	0.78	CeO_2	6.6	β,γ	2×10^{-9}	25.6	3.8	25.3	126	1	Heavy (10.2)	Being considered for short-lived terrestrial heat sources
^{147}Pm	2.6	Pm_2O_3	6.6	β	2×10^{-8}	0.33	0.27	1.8	2788	200–600	Minor (1)	Being used in thermal conditioning applications
^{170}Tm	0.35	Tm_2O_3	7.7	β	1×10^{-8}	12.1	1.07	8.2	445	10–25	Moderate (2.5)	Being considered as alternative to ^{210}Po
^{210}Po	0.38	GdPo	10	α	2×10^{-9}	141	82.4	824	32	10–25	Minor (1)	Reference space short-lived fuel
^{238}Pu	87.4	PuO_2	10	α	7×10^{-13}	0.50	0.36	3.6	30	500–700	Minor (0.1)	Reference space long-lived fuel
^{242}Cm	0.45	Cm_2O_3	9	α,n	4×10^{-11}	120	98	882	28	-	Minor (0.4)	
^{244}Cm	18	Cm_2O_3	9	α,n	3×10^{-12}	2.8	2.5	22.5	29	100–500	Moderate (2)	Being considered to supplement ^{238}Pu

*MPC = maximum permissible concentration.

†For transfers to other federal agencies only, based on estimated costs in the late 1970's.

‡Value in parentheses is inches of lead shielding required for a 1-kw source to reduce radiation to 10 mr/hr at 1 m. Does *not* include neutron shielding for curium.

irradiation, to process specific isotopes after irradiation, or to manufacture isotopic sources.

The cost to produce isotopic source materials depends upon several critical factors, among which the most important are the quantity and quality of the material to be produced, the mixture of isotope products produced, and the relative values of these irradiated products. These factors can influence significantly the mode of operation of production reactors and thus the irradiation and fuel-cycle costs of the various products. Accordingly, it is not possible to quote firm costs which can be applied in a general way to specific isotopes in the absence of firm requirements. The probable ranges of costs to produce in government facilities large quantities of specific isotopes for transfer to other federal agencies have been estimated and are indicated in Table 1.

Potential fission-product availability will grow as the number of commercial power reactors starts increasing in the early 1970's. Within 5 years, commercial fuel-reprocessing plants could supply ^{90}Sr and ^{137}Cs up to the equivalent of 50 kw(t) of each per year and aged ^{147}Pm up to the equivalent of 2 kw(t) per year. However, such production would require the installation of fission-product recovery facilities at these processing sites, and other facilities would be required to manufacture isotopic heat or radiation sources.

Production and distribution of kilocurie quantities of ^{60}Co began in 1951 and since then have grown to megacurie quantities. As private industry capability has developed, the AEC has withdrawn from the production and sale of lower-specific-activity ^{60}Co.[1] Lead times of 12 to 18 months are required for production of high-specific-activity material.

Thulium-170 could be supplied from AEC production reactors in quantities as great as megawatts per year. The availability of ^{170}Tm depends upon the availability of ^{169}Tm target material and high-flux reactors to produce a high-specific-activity fuel form.

Unless a low-cost method for producing ^{170}Er is discovered, ^{171}Tm, which has desirable characteristics for low-shielding heat-source applications, cannot be made in existing production facilities at costs less than several thousands of dollars per watt.[2]

At present, the only capability to process ^{210}Po or manufacture ^{210}Po heat sources exists at AEC's Mound Laboratory, and its encapsulation capability is currently limited to a few hundred grams of ^{210}Po annually. By 1969, however, Mound should have the capability to supply 60 kw(t) of ^{210}Po annually. When requirements for ^{210}Po exceed Mound's capacity, facilities will be required to process annually several hundred tons of irradiated bismuth; recover the bismuth as metal target billets for recycle; and recover, purify, and encapsulate ^{210}Po in the amounts required.

Plutonium-238 is made by the addition of a neutron to ^{237}Np followed by β^- decay. Campaigns to produce large quantities of ^{238}Pu typically require 1 to 5 years of lead time. Starting in the late 1970's, irradiated power-reactor fuels will be the primary source of ^{237}Np supply. Therefore recovery and conservation by the nuclear industry would be a major contribution to the future availability of ^{238}Pu for isotopic power applications. It appears that only modest added capital expenditures would be necessary for ^{237}Np recovery at commercial reactor fuel-processing plants. If the ^{237}Np potentially available from the commercial reprocessing of power-reactor fuels is recovered and accumulated, and then irradiated in government or private reactors to produce ^{238}Pu, the total annual supply of ^{238}Pu can thereby be increased substantially. Neptunium-237, produced as a by-product in power reactors, should be less costly than that produced in AEC reactors because of the higher production rate through longer fuel exposures. A lower ^{237}Np cost, plus cost reductions anticipated in processing and encapsulating techniques, would lead to the lower limit of the ^{238}Pu costs shown in Table 1.

The high power density of ^{244}Cm makes it better than ^{238}Pu for thermionic power using the integral diode-fuel arrangement—the simplest, most straightforward design; however, the neutron dose due to spontaneous fission of ^{244}Cm requires more shielding than ^{238}Pu. Although a considerable amount of investigation using gram quantities is under way, characterization of ^{244}Cm for the required regimes of temperature and metallurgy will not be completed until 1969–1970.

Significant amounts of high-exposure plutonium, americium, and curium isotopes will become available from the nuclear power reactors[3] starting in the late 1970's. This could increase the availability and reduce the lead time and cost to produce large amounts of ^{244}Cm. The cost range of Table 1 is based upon the recovery of ^{244}Cm from spent power-reactor fuel.

Role of Isotopes in National Programs

Systems using radioactive isotopes as sources of heat or radiation are making increasingly important contributions to several vital national goals. Figure 1

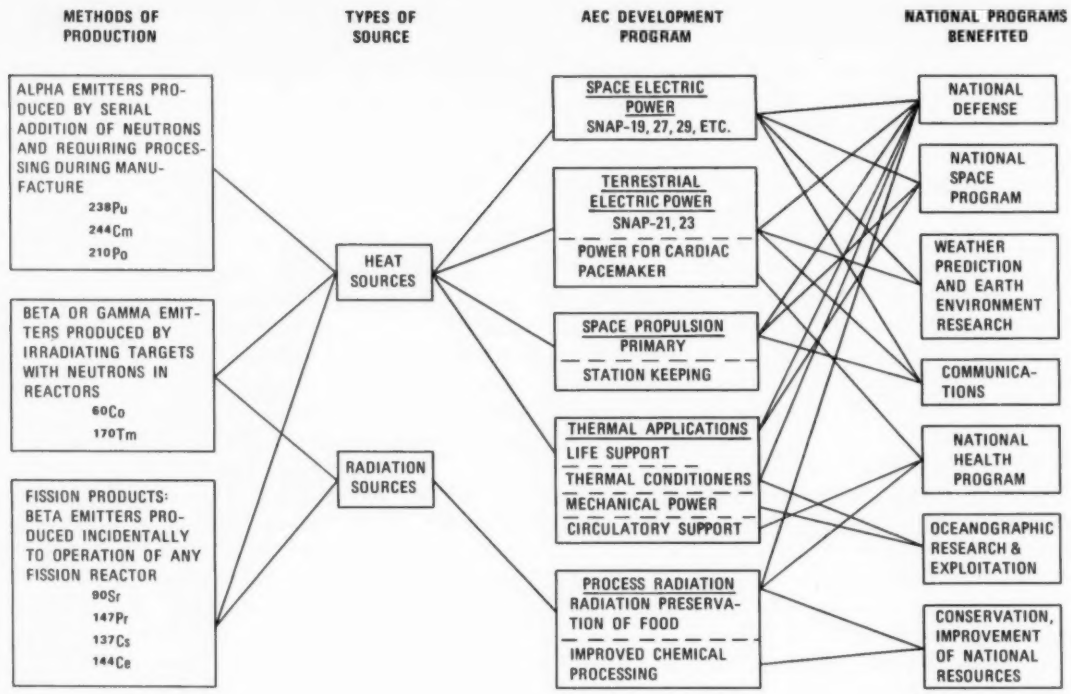

Fig. 1 AEC development of isotope sources for national programs.

illustrates the interrelationships of AEC isotope-development programs and the national benefits to be derived, as well as the types of sources and the methods of production.

Isotopes are in use as heat sources in space and terrestrial electric power, heat, and radiation systems serving the national defense, as well as in other systems which contribute to national security.

In the national space program, isotopes will serve as heat sources for peaceful and military space power systems, propulsion systems, life-support systems, and environmental control systems. Isotope-fueled space power systems are expected to improve the capabilities of satellites for weather prediction and communications as well as for navigation, earth environment research, and space exploration.

In the national health program, isotopic heat sources may be used in systems to power circulatory support systems,[4] as well as to provide electric power for cardiac pacemakers.

In the increasingly important sphere of oceanographic research and exploitation, isotopic heat sources for propulsion, electric power, mechanical power, life support,[5] and swimsuit heaters are expected to play significant roles.

Isotopes as radiation sources are becoming increasingly important in the conservation of national resources through pasteurization, sterilization, and disinfestation of foods, and in chemical manufacturing processes.

Space Power Systems

The isotopic power system currently available for use in space is the radioisotope thermoelectric generator (RTG), fueled with ^{238}Pu (Ref. 6). Its nonnuclear competitor is the solar cell-battery combination, which will continue to be used for a wide variety of missions. (For missions lasting only a few days, primary batteries and fuel cells will continue to be useful.) In some situations the ^{238}Pu-fueled RTG offers advantages over the solar cell system, in particular, where only limited amounts of sunlight are available, where invulnerability to natural and artificial radiation is required, and where the greater compactness, ruggedness, simplicity of integration of the

system, and invulnerability to environmental temperature, available with RTG's, are at a premium. This simplicity of integration includes the availability of large quantities of waste heat to simplify spacecraft environment control. The nuclear competitor is the fission reactor power system, which becomes attractive at power levels in excess of a few kilowatts. Improved performance and lower cost will expand the number of areas of isotope competitiveness and the number of mission applications.

Radioisotope thermoelectric generators have been used successfully[7,8] in two Navy Experimental Navigation Satellites (SNAP-3A, 2.7 watts) and two Navy Operational Navigation Satellites (SNAP-9A, 25 watts). A 50-watt power supply consisting of two SNAP-19 generator modules was launched as part of the Nimbus-B NASA weather satellite in early 1968, and a 56-watt SNAP-27 is to be left on the moon by the Apollo astronauts as part of the Apollo Lunar Surface Experiments Package (ALSEP). The Department of Defense (DOD) has requested AEC to develop a new power unit for the Navy Operational Navigation Satellite (NAVSAT) and has also requested development of the ^{210}Po-fueled, 500-watt, 90-day-life SNAP-29 unit,[9] the latter now in system technology development. NASA has requested AEC to develop and deliver a 25-kw(t) heat source during the 1971 period for use with the isotope–Brayton cycle system to be ground tested at NASA's Lewis Research Center. The NAVSAT and the isotope–Brayton system programs are under discussion with DOD and NASA. In the area of advanced systems technology, a multihundred watt RTG is being designed for possible future missions. This design study affords a convenient means to investigate the safety and ground handling problems of ^{90}Sr (which has potential advantages of economics and availability) in the context of a practical power-system design, in addition to the usual ^{238}Pu design provisions. Advanced technology projects in fuel forms and power conversion systems are being carried out in a coordinated effort with NASA and DOD.

Power systems under development may be divided into several categories. The first category consists of generators for power levels up to about 100 watts(e), incorporating fuel, thermocouples, and integrally mounted waste heat radiators in one package. This category includes the SNAP-19 and SNAP-27 flight systems as well as the 30-watt "lightweight" thermoelectric generator. A second category consists of intermediate-sized RTG's with separately mounted radiators cooling the thermoelectrics. SNAP-29 is an example of this category. A third category consists of power systems in the multikilowatt power range. To conserve isotopes these systems will use highly efficient dynamic power conversion systems. In all categories, power systems composed of building block "modules" are being developed to meet the widest possible combination of potential power requirements. Additional categories or subcategories of systems may be defined in the future, as requirements for these new systems are identified.

Future use of isotopes in space may be paced by the attainment of advanced fuel and system technology, especially as these enable design of higher temperature, more-efficient systems. The most promising approach to increasing the usefulness of isotope systems is to develop higher system efficiency. There are two main avenues of attack:

1. Raising the heat-source temperature capability to increase system efficiency for a given heat disposal radiator area.
2. Developing more-efficient electrical generator concepts, such as cascaded thermoelectrics, dynamic machinery, and thermionic generators. (Many of these advanced generators also require higher heat-source temperatures than are currently available.)

The capability of higher temperatures is one of the most difficult goals of the current isotope fuel form development program. Compared to a current 760°C surface temperature capability, the goals for various propulsion and power conversion concepts are as follows:

Advanced Rankine, Brayton, and thermoelectric generators, and auxiliary thrusters, 980 to 1205°C
Thermionic generators and primary thrusters, 1690 to 2010°C

Figure 2 illustrates the trend of power levels and temperatures in isotopic space power systems.

For repetitive space power applications, there is a strong incentive to use a cheaper long-lived fuel. Strontium-90 is cheaper than the present reference space system fuel, ^{238}Pu, but its radiation shielding and safety problems in space systems are difficult to evaluate. It is not yet clear whether ^{90}Sr can be made safe and convenient enough for space use; investigations are proceeding.

A critical factor in the widespread use of space isotope systems is the resolution of safety assurance standards and the solution of several vital technical problems related to safety, such as methods of fuel encapsulation to ensure integrity in all expected operating and accident conditions.[10,11] The criterion in

all cases is that the isotope devices be operated in such a way that no undue risk to the public occurs. In almost all cases, higher fuel temperature capability could assist in resolving the safety problems.

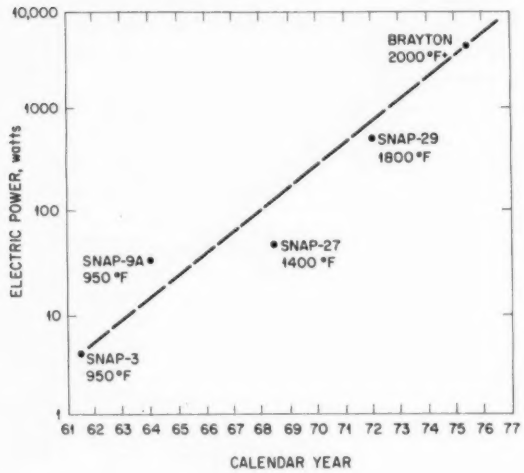

Fig. 2 Trend of power levels and operating temperatures in space power systems.

Because of the substantial financial commitment and several-year lead times needed to produce the isotopes of major interest for space and certain other specialized applications, there is an urgent need for better long-range forecasting of needs by users. Generally, the users, paced by mission funding authorization, forecast their firm requirements no more than 2 or 3 years in advance of required delivery. Development of fuel forms and power conversion systems requires lead times generally in excess of this 2- or 3-year lead in firm requirements information; and for this reason, advanced technology work must be scheduled on the basis of estimates of preliminary requirements. The level of actual demand for isotope systems will, in turn, be determined largely by the degree of success of development programs.

Despite the difficulty of forecasting, NASA has encouraged AEC to produce 500 kw(t) of ^{238}Pu by 1980 to meet the space power needs of the peaceful space program. Meanwhile, quantities of ^{238}Pu forecasted for DOD and NASA and required by current AEC development programs appear now to be approximately equal to all that AEC can produce through about FY 1971.

Terrestrial Power Systems

As with space power, the AEC approach to development of terrestrial power systems is to study classes of potential applications and the factors which affect competitiveness with other energy sources in each class; then, to develop technology and prototype systems which satisfy the competitive criterions, and to cooperate with user agencies in testing in simulated and actual employment situations. In this manner the lowest possible total number of separate development projects is required, and the changing schedules of employment by individual user systems should have the least possible effect on the overall development program.

In terrestrial applications, isotope-system weight is less important than in space systems. Therefore shielding for beta and gamma emitters can be provided, allowing the use of various fission-product fuels which in the future will be available in large quantities as by-products of the civilian central-station nuclear power industry.

In most terrestrial applications, reliability, economic advantage, safety, and portability must be demonstrated to the satisfaction of potential users. Factors advantageous to economic competitiveness of isotopic power sources include high reliability and reproducibility, low operating cost, and long unattended life. For certain unattended surface and undersea applications, the long life of isotopic systems offers a unique operational regime; there are no competitive power concepts. Long-life reliability is also the key to attractiveness in applications such as isotopic cardiac pacemakers.

Proof-of-principle ^{90}Sr RTG's have been developed under the SNAP-7 program and have operated with reasonable success at several terrestrial sites[12,13] including remote unattended weather stations, a navigation buoy, an undersea research experiment, and an off-shore oil rig. Advanced systems, which will capitalize on this SNAP-7 experience to meet evolving economic and reliability objectives, are being developed for marine (SNAP-21) and land (SNAP-23) applications in the early to middle 1970's. Plutonium-238-fueled microwatt and milliwatt generators for cardiac pacemakers and low-power electronics power sources are also being developed. Cobalt-60, ^{144}Ce and ^{147}Pm are being evaluated as fuels for terrestrial power systems, in addition to ^{90}Sr. AEC hopes in the future to initiate an applications engineering and parametric design study of isotopic heat sources and dynamic

conversion equipment in the 1- to 20-kw(e) power range for various terrestrial and undersea needs.

In conjunction with specific system-oriented work, the terrestrial power program also includes several projects for improvement of basic technology common to one or more current and future generator concepts. These include development of better thermal insulation, improved components of thermoelectric and other electrical generators, improved fuel forms, and studies of methods of safe design and employment of isotopic systems.

Thermal Applications

Thermal conditioning systems such as spacecraft and swimsuit heaters, which may be lighter and more reliable than electrically powered heaters, are being investigated. Isotope-fueled life-support systems appear attractive for long-duration space missions when compared with solar energy. In this category a heat source for a waste-water reprocessing cycle has been demonstrated for manned space applications.[5]

Thermodynamic systems being considered include an energy source for circulatory support devices. The technical problem is an extremely challenging one; and, if successfully developed, this application alone could conceivably use all foreseen supplies of ^{238}Pu. Promethium-147 is also being considered as a candidate fuel. Design studies to define an isotope-powered engine concept for a heart assist or replacement are being performed in cooperation with the National Heart Institute.

Process Radiation Systems

Isotopes are in use as radiation sources in several industrial processes where this radiation is more economical than nonnuclear processes or accelerators used as radiation sources, or where unique products can be produced.

Economic competitiveness is a key criterion even in situations where radiation produces a better product, since the margin of improvement is usually amenable to some form of economic analysis. In some cases, however, such as radiation-processed wood–plastics, a final estimate of the value of the new product must await gradual acceptance or nonacceptance by the commercial market.

New and expanded uses for radiation processing are continually being investigated. Several radiation-processed materials have been developed to the point of active industry investment.[14-20] Processes for radiation pasteurization of foods are being developed for eventual commercialization by developing the technology through semicommercial scale demonstrations, and obtaining Food and Drug Administration clearance on a limited number of foods for which such processing appears to offer substantial economic benefit.[19,21-23]

Space Propulsion

Radioisotope-heated thrusters use the thermal energy produced by the decay of an encapsulated radioisotope to heat a propellant, which is exhausted through a rocket nozzle to produce thrust. These thrusters can provide relatively low thrust (10^{-6} to 10^{-1} lb) and high-specific impulse (200 to 800 sec, depending on propellant selected and capsule temperature capabilities) in a simple, lightweight engine.

Two general classes of radioisotope-heated thrusters have been under investigation: one is a high-temperature (1650 to 1925°C) high-specific-impulse (750 to 800 sec), 0.25-lb-thrust, high-power [5 kw(t)] class which uses hydrogen propellant for primary propulsion application. The other is a lower temperature (980 to 1205°C) low-thrust (micropound to millipound), low-power (100 watts) class which, using ammonia propellant, provides specific impulse in the range of 200 to 250 sec for auxiliary spacecraft propulsion. Work on the high-temperature, primary propulsion thruster has been suspended pending development of the necessary high-temperature fuel form and capsule technology.

The Air Force and the AEC are currently considering a cooperative flight test program of an auxiliary radioisotope-heated thruster using ammonia propellant to demonstrate flight hardware and make development lead times compatible with spacecraft development schedules; but no specific mission requirement has been stated, and no flight program has been formulated.

Outlook

AEC will continue to be faced with important technical and administrative problems in encouraging effective use of fission products and other isotopes, forecasting future production requirements, and ensuring availability of desirable isotopes. Nevertheless, important progress has been made in the first years of isotope development, and a future of ever-increasing value of isotopes to the national interest is to be expected.

Production

Background

The planning for isotopes production involves several considerations. Continued long-term planning and analysis are required to select those products which will make best use of available government facilities, as well as those which will provide technology needed to take best advantage of private production capability as it becomes available.

One vital area for consideration is the utilization of the fission products ^{90}Sr, ^{137}Cs, ^{144}Ce, and ^{147}Pm. At present these fission products represent a significant liability to the operators of commercial power reactors. One of the objectives of the AEC isotopes programs is to develop the technology to use fission products economically, taking advantage of their value as heat and radiation sources so that they will be an asset rather than a liability to the national economy.

Requirements. Long-Lived Isotopes (^{238}Pu, ^{244}Cm, ^{90}Sr). The systems (described later) which are under research and development or scheduled for delivery to users will require about 10 kg of ^{238}Pu per year in FY 1968–1970; these types of systems are estimated to require about 60 kg per year in FY 1971–1974 and 120 kg per year in FY 1975–1977.

Comparisons of requirements with production capability indicate that it is possible to meet current firm requirements for ^{238}Pu but that potential levels of demand through 1980 may exceed the total of AEC and private power-reactor production capacity. No excess is foreseen through 1971, at least. Throughout the second half of the 1970's, there could be a net cumulative deficiency of production compared to requirements.

Many requirement forecasts for long-lived isotopes assume the use of ^{238}Pu because of its current state of development and its desirable radiation characteristics. Some alternative isotope should be made available to supplement the production and to permit ^{238}Pu to be used in applications where no other isotope will serve as well. The characteristics of ^{244}Cm are superior in some respects to ^{238}Pu; but ^{244}Cm has not been available, and the technology needed for its use has not been developed. The use of ^{244}Cm probably will be limited in some applications by the need for shielding from the radiations emitted due to spontaneous fission. Similar shielding problems exist for ^{90}Sr, and these problems are further compounded by the potential biological hazard of available ^{90}Sr fuel forms.

Curium-244 may be preferable to ^{238}Pu in thermionic power conversion devices because of its higher specific power and indicated capability to maintain a higher temperature at the emitter of a thermionic conversion device.

The requirements for ^{90}Sr have been associated mostly with terrestrial applications. The cumulative demand for ^{90}Sr for terrestrial power uses is estimated to be approximately 75 kw(t) with a range of 35 to 90 kw(t) by 1975, and 125 kw(t) with a range of 60 to 150 kw(t) by 1980. Additional quantities for thermal and radiation applications could be as high as 300 kw(t) by 1975. Supplies of ^{90}Sr seem to be adequate to meet current demands, but a processing plant will be required for separation and encapsulation when more than about 10 kw(t) per year is required.

Short-Lived Isotopes (^{210}Po, ^{147}Pm, ^{170}Tm, ^{60}Co). The only firm requirement for ^{210}Po is 0.25 kg for the development of a prototype SNAP-29 generator of about 28 kw(t). Additional research and development may require about 0.5 kg per year. Use of ^{210}Po by NASA and DOD in various power devices could result in a total requirement of about 6 kg per year in FY 1972–1977.

Firm requirements for ^{147}Pm in FY 1967–1968 are relatively small—65 watts(t) for an Air Force Inertial Guidance Heater, 50 watts(t) for a microthruster, and 500 watts(t) for research and development (R&D). In subsequent fiscal years through 1970, 1 kw(t) per year will be required for R&D. The potential Air Force heater requirement is 13 kw(t) per year, beginning in 1969.

Only developmental quantities of ^{170}Tm are currently projected as a requirement. For possible demand levels, ^{170}Tm may be assumed to be an alternative to ^{210}Po.

Cobalt-60 is used mainly for radiation processes and is scheduled for evaluation in heat and power sources. Cobalt-60 is supplied in a wide range of specific activities, from a few curies per gram to hundreds of curies per gram, depending on the application. Requirements are currently several kilograms (several megacuries) per year, and may average about 10 kg per year in FY 1969–1972 and about 30 kg per year in FY 1973–1977.

Cost and Availability Projections. The basic reactor, process, and source-encapsulation technology for producing radioisotope sources exists, and it is being improved. The program also included plans to demonstrate the technology required for production of large quantities of special isotopes at reasonable costs.

Assuming that requirements for defense products would permit space in AEC reactors to be used for radioisotope production, the availability of important radioisotopes in large quantities is currently limited by the lack of facilities necessary to prepare targets for irradiation, to chemically process specific isotopes, or to manufacture isotopic sources. These facilities would be specific to the product or products to be manufactured. A decision to build these facilities depends on the justification (demand, and benefit or value) to support the capital investment required.

Factors Influencing the Major Cost Components. The irradiation cost to produce isotope source materials depends upon several critical factors, among which the most important are the amount and specification of the material to be produced. Other important factors are the total isotope product mix produced and the relative values of these irradiation products. These factors can influence significantly the mode of production-reactor operation, and the irradiation and fuel-cycle costs of the various products. Accordingly, it is not possible to quote firm costs which can be applied, in a general way, to specific isotopes; therefore a cost goal range within which the costs for specific isotopes may fall has been chosen for general use.

The influence of several other factors can be seen from a brief discussion of the ways they affect the cost of an important neutron product, ^{238}Pu. The cost of an encapsulated ^{238}Pu heat source includes the following components:

1. Cost of ^{237}Np and target fabrication.
2. Irradiation costs.
3. Separation of plutonium from neptunium.
4. Recovery and preparation of ^{237}Np targets.
5. Unrecoverable waste (losses) and ^{237}Np inventory cost.
6. Conversion to PuO_2 fuel form and waste recovery.
7. Encapsulation.

There will be significant quantities of ^{237}Np produced as a by-product in nuclear power-reactor fuels, and it is expected that ^{237}Np recovered during irradiated fuel processing will be procurable at a negotiated price more favorable than the cost of ^{237}Np produced in AEC production reactors.

Because of the inherently slow rate of conversion of ^{237}Np to ^{238}Pu, the ^{237}Np inventory may tie up a substantial amount of money; and management of this inventory may have a substantial effect on ^{238}Pu costs. The cost of conversion of the ^{238}Pu, once separated from the ^{237}Np target, into a usable PuO_2 fuel form, including recovery of the wastes which result from the process, accounts for almost 30% of the cost of the final ^{238}Pu heat source. The high current costs result largely from the early developmental status of the process for manufacture of PuO_2 microspheres, which is being done on a laboratory scale. A new process now under development promises to reduce this cost of making microspheres to about one-fifth of its present value. Finally, the unit cost of PuO_2 fuel form encapsulation is expected to be reduced by at least half due to mass production economies when output increases to the production rates expected by the mid 1970's. It is this type of cost variation which leads to the wide range of ^{238}Pu cost goals in this article.

As regards fission products, AEC cost goal ranges are based on 1963–1965 design studies of fission-product processing plants, normalized to the stated assumptions. Fission-product costs are very sensitive to the total market for all fission products. For example, ^{90}Sr, ^{137}Cs, and ^{144}Ce must be separated from each other and from the rare earths. The rare earths must be processed along with ^{147}Pm, and the costs of operation of common facilities must be charged off against the total market, even if only one of the isotopes is needed. The unit cost is therefore also heavily dependent on production quantities, varying almost inversely with output.

Basis for Preliminary Manufacturing Cost Projections.* It is difficult to project manufacturing costs for supplying radioisotopes without considering the amounts and specifications of the material required and other relevant considerations which relate time, money, and human resource requirements. However, cost information can be a starting point to permit a potential user to determine whether the use of a radioisotope may be of potential value, or which radioisotope might have greater value. Accordingly, manufacturing cost goals for the major radioisotopes have been developed which reflect what might be achievable for large quantity isotope production in government facilities. Although these cost goals reflect primarily the relative costs of alternative isotopic energy sources, they may also be used to estimate what the potential costs to federal agencies might be if large amounts of these specific isotopes were supplied from facilities operated by the AEC.

*Unless specifically noted, manufacturing costs as used herein are for the entire process sequence from target procurement and preparation for irradiation through delivery of the radioisotope in a primary containment capsule.

Operations beyond primary capsule containment require certain handling, welding, measurements, and quality assurance testing that vary with the application. Since the final encapsulation cost could vary from source to source depending upon size, complexity of design, materials of construction, and fabrication or quality assurance specifications, this cost has not been included in the manufacturing cost goals.

Fission-Product Prices. At present, fission products in annual quantities up to the amounts shown are available from the AEC at the following prices:

	Mc/year	Cents/curie	$/thermal watt
^{90}Sr	5–10	20	30
^{137}Cs	2	12½	26
^{147}Pm	6	20	560

These prices are for bulk, unencapsulated products in large quantities and were established to stimulate use. They are based on a study made to determine the probable costs for these isotopes if produced in large quantities in a hypothetical commercial facility at Hanford.[24]

Manufacturing Cost Goal Ranges. The following cost goal ranges have been estimated for large quantities for use by federal agencies. These intragovernment charges do not include several factors such as depreciation and profit, which must be included in commercial prices.

Isotope source	Manufacturing cost, goal range–$/thermal watt
^{90}Sr	25–35
^{137}Cs	20–30
^{147}Pm	200–600
^{60}Co	7–25
^{170}Tm	10–25
^{210}Po	10–25
^{238}Pu	500–700
^{244}Cm	100–500

These estimates assume that the isotopes are produced, processed, converted to a fuel form, and sealed in a primary containment capsule in government-operated facilities. The cost to chemically process target materials and to convert a product to a fuel form in large quantities is small in comparison to the costs for certain target materials and target irradiation. However, the bulk of the capital and fixed operating costs of a new large processing and manufacturing facility are required even for small quantities; thus the unit cost of processing and source manufacture for small quantities may be the major cost element in the production of a specific isotope source, especially if irradiation costs are shared by other products.

Objectives

Within the framework of the overall AEC objectives for the isotopes program, and recognizing the role of private industry, the production program objectives are as follows:

1. Develop new technology for production of reactor products in existing AEC and private production facilities to assure a continuing return on the large capital investment they represent.
2. Maintain viable facilities for national defense and security.
3. Produce the required quantities of those isotopes which can be produced best in AEC production facilities.

Program Plan

Production Techniques and Capabilities.* The following are primary considerations which influence the availability of the major radioisotopes.

Fission Products. Fission products are formed in reactor fuel elements when ^{235}U, ^{233}U, and ^{239}Pu fission. These by-products can be recovered from the waste streams which result from the chemical reprocessing of reactor fuel. Their availability is a function of the fuel exposure and the quantities of nuclear fuel reprocessed. Processes for the recovery, separation, and purification of fission products have been developed, and they are being improved. At present, a few megacuries annually of ^{90}Sr, ^{137}Cs, and ^{147}Pm can be processed at Hanford, and converted to fuel forms and encapsulated at the Oak Ridge National Laboratory.

Under the AEC's Waste Management Program at Hanford, an appreciable quantity of the long-life,

*Papers on "Large Scale Production and Applications of Radioisotopes" were presented at the American Nuclear Society's National Topical Meeting held on March 21–23, 1966, in Augusta, Georgia. The proceedings of this meeting were published in Report DP-1066, Vols. I and II, available from the Clearinghouse for Federal Scientific and Technical Information, National Bureau of Standards, U. S. Department of Commerce, Springfield, Va. 22151.

heat-producing fission products ^{90}Sr and ^{137}Cs will be removed from both current and stored chemical reprocessing plant wastes so that the residual waste may be solidified for long-term storage. The ^{90}Sr and ^{137}Cs crude fractions which will be removed from the wastes at Hanford will be temporarily stored as a nitrate solution.

Beginning in 1968, tens of megacuries annually of ^{90}Sr and ^{137}Cs could be made available from the Hanford crude fractions. Although ^{147}Pm is not removed for waste management, a crude ^{147}Pm fraction also could be made available during waste processing operations. A facility capable of purifying such large quantities of ^{90}Sr, ^{137}Cs, and ^{147}Pm and manufacturing encapsulated sources from the fission products at Hanford was estimated to cost about $10 to $15 million if built at Hanford and operated in conjunction with the waste management program. However, it may be possible to recover large quantities of specific fission products with a lower capital investment, and still meet projected manufacturing cost goals.

Small commercial fuel-reprocessing plants, capable of annually processing 300 tons of nuclear fuel exposed to about 25,000 Mw days per metric ton, could supply ^{90}Sr and ^{137}Cs in amounts up to the equivalent of 100 kw(t) of each per year and aged ^{147}Pm up to the equivalent of 5 kw(t) per year. However, additional facilities would be required to recover these products from the chemical reprocessing wastes and to manufacture isotopic sources. Several fission-product processing facilities at different processing sites would be required to handle the substantial amounts of the fission products that will be contained in irradiated fuel from the growing nuclear power industry.

Reactor-Produced, Nonprocessed Isotopes. *Cobalt-60.* This isotope is made by addition of a neutron to readily available mononuclidic cobalt (^{59}Co). Production and distribution of kilocurie quantities of ^{60}Co began in 1951 and have grown to megacurie quantities. The AEC has a capability for producing hundreds of megacuries of ^{60}Co. This is far more than the foreseeable requirements. In May 1961 the AEC withdrew from the production and distribution of ^{60}Co of greater than 30 curies per gram specific activity (modified in 1965 to greater than 45 curies per gram) in view of industry capability to serve routine commercial applications such as teletherapy and radiography. This has led to the development of an industry which encompasses ^{60}Co producers and source encapsulators. The industry is viable; and as new markets for ^{60}Co develop, additional private participation can be expected. For example, ^{60}Co is now being produced in one private nuclear power reactor.[25] The Commission is currently considering withdrawing from the production and distribution of ^{60}Co sources of 45 curies per gram specific activity and less, in light of private capability to produce such material. It is also reasonable to expect that industry may provide ^{60}Co source-encapsulation services for isotopic heat-source applications as well as for radiation applications.

The user desiring large quantities of high-specific-activity ^{60}Co must allow a lead time of about 12 to 18 months or more for irradiation and encapsulation of ^{60}Co, depending upon the reactor flux available and the specific activity of the material to be produced.

Thulium-170. This isotope is made by addition of a neutron to ^{169}Tm, a rare earth. Because of its short half-life (127 days), the user must allow for decay of the ^{170}Tm, e.g., about 10% between reactor discharge and delivery of the target assembly, and an additional 4% per week during manufacturing of the encapsulated sources and assembly of the power unit for delivery. Thulium-170 could be supplied from AEC production reactors in up to megawatt quantities per year. The availability of ^{170}Tm depends upon the availability of ^{169}Tm target material, high flux levels to produce a high-specific-activity fuel form, and facilities to convert the irradiated targets into a usable isotopic source.

Thulium-170 was the first isotope produced in kilocurie amounts in a private reactor. In 1959, General Electric produced approximately 150,000 curies of ^{170}Tm, which was used to power an experimental thermionic generator. Multikilowatt quantities of ^{170}Tm could be produced in existing commercial facilities on relatively short notice.

Thulium-171. This isotope has frequently been considered as a desirable isotopic source because of its 1.9-year half-life and minimal shielding requirement. There are several ways to produce ^{171}Tm, but all are prohibitively expensive. There is no known economic method for isotopic enrichment to produce the multiton quantities of enriched ^{170}Er needed to make ^{171}Tm in large amounts. Unless a low-cost method for producing ^{170}Er becomes available, ^{171}Tm cannot be made in AEC production facilities at costs less than several thousands of dollars per watt.[2]

Reactor-Produced, Processed Isotopes. *Polonium-210.* This isotope is made by addition of a neutron to

readily available mononuclidic ^{209}Bi and subsequent decay. Because of the very low cross section for the reaction (0.019 barn), many tons of bismuth must be irradiated and processed to produce ^{210}Po in large quantities. Because of the short half-life (138 days), the user must allow for decay of the ^{210}Po after its discharge from the reactor. Furthermore, since its recovery cost is related to the amount of bismuth which must be processed, it is preferable to irradiate the bismuth at high flux levels to obtain as high as possible a concentration of ^{210}Po in the bismuth target discharged from the reactor.

At present, there is no commercial capability to process ^{210}Po or manufacture ^{210}Po heat sources. Such capability exists only at Mound Laboratory, and its encapsulation capability is currently limited to a few hundred grams of ^{210}Po annually. By 1969, however, its process facilities will have been improved, and a new facility for encapsulating ^{210}Po will have been completed. Mound Laboratory will then have the capability to supply 1.0 to 2.0 kg of ^{210}Po annually.

Plutonium-238. Plutonium-238 is made by the addition of a neutron to ^{237}Np followed by β^- decay. Neptunium-237 is a by-product formed in a reactor fuel element by (1) addition of a neutron to ^{236}U which may be formed in about 20% of the neutron captures by ^{235}U, and (2) an $(n,2n)$ reaction with ^{238}U.

Since ^{237}Np is a by-product of nuclear power generation, any increase in planned nuclear power generating facilities would increase the amount potentially available from that source. If the ^{237}Np expected to be available from the commercial reprocessing of power-reactor fuels is recovered and accumulated, and then irradiated in government or private reactors to produce ^{238}Pu, the total annual supply of ^{238}Pu in the late 1970's could more than double that produced from AEC ^{237}Np. At these higher production rates, limited AEC equipment modifications would be required for ^{237}Np target fabrication, for processing irradiated ^{237}Np targets, and for conversion of the ^{238}Pu to an oxide. Recovery from irradiated power-reactor fuel, with only modest added capital expenditures, at commercial reprocessing plants, will be the primary future source of ^{237}Np supply. Its recovery and conservation by the nuclear industry would be a major contribution to the future availability of ^{238}Pu for isotopic-power applications.

Figure 3 projects the ^{237}Np potentially available annually from spent power-reactor fuels. The curve is based upon a projected U. S. nuclear power capacity in

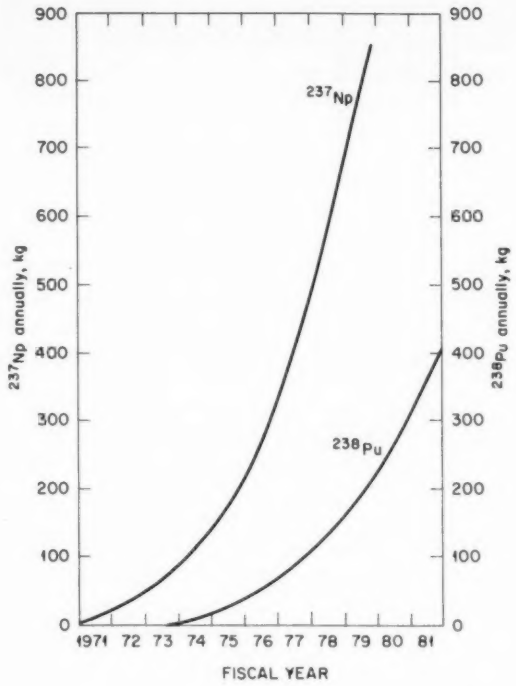

Fig. 3 ^{237}Np and ^{238}Pu potentially available from power reactors.

1980 of 145,000 Mw(e). The ^{237}Np projection ignores the effects of recycling uranium and plutonium, but allows 1 year from spent-fuel discharge to availability of ^{237}Np for target fabrication. The ^{238}Pu availability curve is based upon the following assumptions:

1. All ^{237}Np would be recovered and recycled for irradiation in reactors to produce ^{238}Pu.
2. The production of 1 kg ^{238}Pu consumes 1.5 kg ^{237}Np.
3. The ^{237}Np-to-^{238}Pu conversion-cycle time would be 1 year, and about 160 g of ^{238}Pu would be produced annually per kilogram of ^{237}Np irradiated.

The General Electric Company has announced that ^{237}Np will be recovered in its fuel-processing plant and offered for sale. The New York State Atomic and Space Development Authority has revealed plans for recovering neptunium in the Nuclear Fuels Service fuel-processing plant. The United Kingdom and Eurochemic have also offered ^{237}Np for sale or barter to the United States.

The AEC has made known its plans that, to the extent procurement of some neptunium will be required, AEC would annually negotiate terms for

delivery of neptunium to AEC as soon as a commercial neptunium-recovery capability is available. The purchases would be contingent upon the successful negotiation of reasonable purchase prices. At the present time, no industry capability exists for producing large quantities of ^{238}Pu from the ^{237}Np; this operation could be carried out in AEC facilities.

An important consideration is that the ^{237}Np, which is part of the "waste products" of the power-reactor fuel-processing cycle, should be recovered during fuel processing and stored. The cost of recovery at this stage is much lower than it would be after the waste is put into long-term storage.

Curium-244. In 1969, AEC will have produced and purified about 5.5 kg of ^{244}Cm for development of a fuel form and for evaluation of this isotope for potential isotopic-power applications.

This isotope is made by addition of a neutron to ^{243}Am, which, in turn, is made by addition of a neutron to ^{242}Pu. Plutonium-242 is made by sequential addition of a neutron to the isotopes of plutonium. If high-exposure plutonium is available as starting material, the production of ^{244}Cm is facilitated.

Significant amounts of high-exposure plutonium, americium, and curium isotopes will become available from the nuclear power reactors. This could increase the availability of target material and reduce the lead time and cost to produce large amounts of ^{244}Cm.

The current production of ^{244}Cm requires a substantial initial investment for the plutonium target material and its initial irradiation (1 year) to produce a subsequent target material containing the ^{242}Pu isotope. It in turn must be irradiated (1.5 years) and then chemically processed to purify and recover ^{242}Pu and ^{243}Am for recycle through the reactor, and to recover the ^{244}Cm product. If large requirements develop, it will be necessary to construct the production facilities required to chemically process large quantities of the irradiated target for recovery of the residual ^{242}Pu, ^{243}Am, and ^{244}Cm; to manufacture target sources from the recovered ^{242}Pu and ^{243}Am; and to manufacture ^{244}Cm encapsulated sources. The manufacturing cost and goal range of this report is based upon recovery of ^{244}Cm from spent power-reactor fuels.

Isotopes Offered for Sale. A catalog of isotopes offered for sale by AEC is available from the Isotopes Sales Department, Isotopes Development Center, Oak Ridge National Laboratory, P. O. Box X, Oak Ridge, Tennessee 37830.

Encouragement of Private Production. As a matter of policy, the AEC has withdrawn from supplying approximately 40 radioactive materials and services, and the AEC will continue to review those services which should be available from commercial sources.

In keeping with this policy, AEC has encouraged private industry to consider (1) making available isotopic materials such as ^{237}Np, (2) providing irradiation services to produce isotopic materials such as ^{60}Co, and (3) the recovery and manufacture of fission-product sources. (PSB)

References

1. AEC Withdraws from Production of Low-Specific-Activity ^{60}Co, *Isotop. Radiat. Technol.*, 5(3): 262 (Spring 1968); 5(4): 347 (Summer 1968).
2. W. H. Pennington, D. W. Cole, and W. E. Mott, Radioisotopic Power Sources for Human Implantation, *Isotop Radiat. Technol.*, 5(4): 312 (Summer 1968).
3. Glenn T. Seaborg, Mass Production and Practical Applications of Actinide Elements, *Isotop. Radiat. Technol.*, 6(1): 1-18 (Fall 1968).
4. See Ref. 2, p. 307.
5. C. A. Metzger, A. B. Hearld, B. J. Reynolds, R. Shivers, and R. W. Murray, Integrated Waste-Management System with a Vacuum Distillation–Vapor Pyrolysis Radioisotope-Fueled Water-Reclamation Function, *Isotop. Radiat. Technol.*, 5(3): 242-251 (Spring 1968).
6. Savannah River Laboratory, Large Scale Production and Applications of Radioisotopes, Proceedings of the American Nuclear Society National Topical Meeting, March 21–23, 1966, Augusta, Ga., USAEC Report DP-1066(Vols. I and II) (CONF-660305), May 1966. See also Ref. 9.
7. International Study of Terrestrial Applications of Radioisotopic Power Sources, *Isotop. Radiat. Technol.*, 4(2): 172-182 (Winter 1966–1967).
8. E. E. Fowler, Radioisotopes and Radiation Developments in the USA—1967, *Isotop. Radiat. Technol.*, 5(3): 176 (Spring 1968).
9. Large Scale Production and Applications of Radioisotopes, *Isotop. Radiat. Technol.*, 4(2): 167 (Winter 1966–1967).
10. Glenn A. Fowler, Aerospace Safety of Isotopic and Reactor Power Sources, *Isotop. Radiat. Technol.*, 2(4): 350-355 (Summer 1965).
11. See Ref. 9, pp. 169-170.
12. ^{90}Sr Powers Navigational Aids in Lighthouse, Atlantic Ocean, *Isotop. Radiat. Technol.*, 2(1): 94-95 (Fall 1964).
13. See Ref. 7, pp. 178-180.
14. Charles Artandi and Walton Van Winkle, Jr., Comparison of Electron-Beam and Gamma-Irradiation Plants, *Isotop. Radiat. Technol.*, 2(4): 321-328 (Summer 1965).
15. Recent Developments in the Production of Wood–Plastic Combinations, *Isotop. Radiat. Technol.*, 3(2): 115-146; 164-166 (Winter 1965–1966).
16. E. Alfred Burrill, Particle Accelerator Uses by Industry, *Isotop. Radiat. Technol.*, 3(4): 297-311 (Summer 1966).

17. Meyer Steinberg, Chemonuclear and Radiation Chemical Process Research and Development, *Isotop. Radiat. Technol.*, 4(2): 142-155 (Winter 1966–1967).
18. David E. Harmer, Radiation Processing: Where We Stand Now, *Isotop. Radiat. Technol.*, 4(3): 265-270 (Spring 1967).
19. See Ref. 8, pp. 172-175.
20. Union Carbide Goes Ahead with Radiation-Treated Polyethylene Oxide, *Isotop. Radiat. Technol.*, 5(3): 266 (Spring 1968).
21. Potential Economic Impact of Commercial Radiation Preservation of Food, *Isotop. Radiat. Technol.*, 3(1): 34-48 (Fall 1965).
22. SINB and USAEC Hold Food-Irradiation Briefing for Industry at Oak Ridge, *Isotop. Radiat. Technol.*, 4(3): 271-274 (Spring 1967).
23. Radiation Preservation of Foods, *Isotop. Radiat. Technol.*, 5(1): 78-82 (Fall 1967).
24. C. A. Rohrmann, Hanford Isotopes Plant, *Isotop. Radiat. Technol.*, 2(2): 99-123 (Winter 1964–1965).
25. Neutron Products Markets First Private ^{60}Co, *Isotop. Radiat. Technol.*, 5(2): 161 (Winter 1967–1968).

American Nuclear Corporation Emphasizes Irradiators

Since its formation in 1961, just outside of Oak Ridge, Tenn., the American Nuclear Corporation (ANC) has specialized in providing ^{60}Co- and ^{137}Cs-fueled radioisotope irradiators, which are now being distributed on a worldwide basis. Two recently sold devices are the Salk and Battelle irradiators mentioned elsewhere in this issue (see page 220). In addition, the company manufactures the RAY-D-TEC line of gamma-radiation-monitoring systems.

Gamma-irradiation services, health-physics services, and irradiator design and consulting services are also provided by the ANC staff, which includes health physicists, design engineers, and technical radioisotopes-oriented personnel.

Facilities

The facility includes approximately 12 thousand sq ft of floor space, of which ~2000 sq ft is allocated to office space, ~3000 sq ft to the hot-cell area, and the balance to the production area and machine shop. The hot cell is capable of handling up to

1×10^6 curies of ^{60}Co and 3×10^6 curies of ^{137}Cs at any one time. Objects up to 6 by 8 by 8 ft can be irradiated at dose rates depending upon source loading (usually ~100 thousand curies of ^{60}Co and ^{137}Cs).

Commercial Products

Medical Sources

For the medical profession, ANC manufactures ^{60}Co wire of various lengths and diameters for use in applicators, ovoids, needles, tubes, and cells, or as sutures; gold-sheath needles; and stainless-steel source capsules. It also provides a complete teletherapy loading and reloading service for all makes of teletherapy sources and makes the sources themselves.

Self-Contained Irradiators

Several types of self-contained mobile irradiators are manufactured by ANC. These are particularly suitable for classroom demonstrations and research irradiations. One of the units (Fig. 1) was used at the

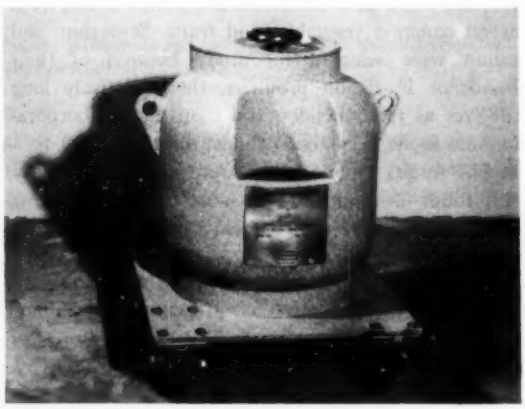

Fig. 1 CES-I-RAD self-contained 1000-curie irradiator.

1964 National Boy Scout Jamboree at Valley Forge, Pa., to irradiate samples of barley seeds for the Scouts.

The CES-I-RAD (^{137}Cs) and CO-RAD (^{60}Co) series provide a wide range of dose rates, depending on the source size. A ^{137}Cs liquid-sample irradiator utilizes an annular source that permits a uniform, concentrated gamma dose for liquid and/or solid samples. A gravity-flow grain irradiator has recently been designed (Fig. 2).

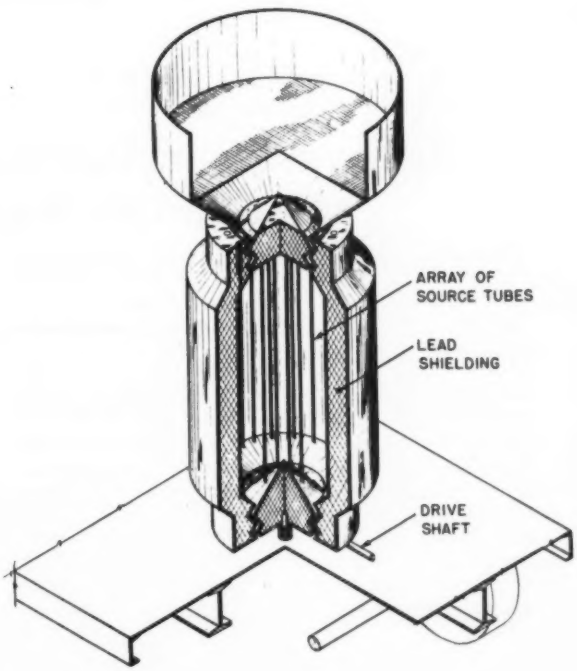

Fig. 2 Gravity-flow grain irradiator.

The GAMMA-CAL units are calibration-beam devices suitable for calibrating large irradiators, with one modification designed to permit irradiations as well as calibrations.

Monitoring Systems

The RAY-D-TEC systems include both portable and stationary devices for monitoring gamma rays, with the former adaptable to alpha- and beta-ray detection, and the latter capable of multichannel installation and hence multistation monitoring. (PSB)

Section

II

Isotopes and Radiation Technology

Isotope Technology Development

Removal of Radioactive Strontium and Cesium from Certain Vegetables and Fruits During Normal Preparation for Preservation*

By J. W. Ralls, H. J. Maagdenberg, T. R. Guckeen, and W. A. Mercert

Abstract: *Spinach, peas, and broccoli were sprayed with a sufficient amount of very dilute solutions of ^{89}Sr or ^{134}Cs, to provide ~1 μc per crop container. Dwarf pear and peach trees were sprayed with slightly stronger solutions to give 4 to 6 μc per tree. Both fruits and vegetables were grown in soil treated with 15 μc of radionuclide for each 40 lb of soil. External contamination was readily washed from fruits and vegetables picked 1 or 2 hr after treatment. Internal contamination could be reduced greatly by ordinary food-preparation processes, except for strawberries.*

To provide adequate nutrition for survivors of catastrophic events producing environmental pollution from radioactivity, food grown on contaminated land may have to be used. In such an emergency, the radioactivity in the edible portion of the crop must be low enough (either when harvested or after decontamination) so that it can be consumed without injury.

This report is concerned with the uptake, distribution, and removal of strontium and cesium in and from several common vegetables and fruits. Strontium and cesium were selected for study because of their abundance in fission products, their relatively long half-lives as radionuclides, and their facile incorporation into foods, bone, body fluids, and tissue.

The results reported here are concerned primarily with foods contaminated with radionuclides of low radiation intensity, which could cause injury only if consumed (without decontamination) for lengthy periods of time. If, for example, a crop was contaminated at a level of 2×10^{-10} to 2×10^{-9} curie in a kilogram portion of prepared food to be eaten frequently, a 90% reduction in the level of radioactivity during preparation would put the residual activity in the Range II Scale of Action of the Federal Radiation Council recommendation[1] for ^{90}Sr intake.

Methodology

The methods used (Fig. 1) to prepare crop samples contaminated externally or internally with ^{89}Sr or ^{134}Cs, and their subsequent treatment under simulated commercial food-preparation conditions, will be outlined in this section. Details on all of the experimental work are in the final report submitted to the contracting agency.[2]

For the study of the problems associated with external contamination, vegetable crops were sprayed

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†Western Research Laboratory, National Canners Association, Berkeley, Calif. 94710.

Fig. 1 Equipment and technique used in spraying crops with simulated fallout.

at or near harvest with a simulated fallout suspension that provided 1 μc of ^{89}Sr or ^{134}Cs for each crop container. Dwarf fruit trees were sprayed with an amount of simulated fallout that contained 4 to 6 μc of the radioisotopes. The crop samples were harvested 1 to 2 hr after spraying and then were prepared by simulating the normal operations used commercially prior to preservation by dehydration, freezing, or canning.

The crop samples used in the study of problems associated with internal contamination by strontium and cesium radioisotopes were grown in soil which had been treated with 15 μc of ^{89}Sr or ^{134}Cs for each 40 lb of soil.

For both externally and internally contaminated experimental crops, samples taken at various stages of the preparation for preservation were analyzed for radioactivity by suspension-gel liquid scintillation spectrometry. The sample was ashed in a muffle furnace (at 760°C) on two filter papers and a bottom layer of 800 mg of Cab-O-Sil. After cooling, 20 ml of a scintillation solution [prepared by dissolving 7.5 g of 2,5-diphenyloxazole and 0.15 g of 1,4-*bis*-2-(5-phenyloxazolyl) benzene in 2 liters of toluene] was added.

The sample was mixed, transferred to a counting vial, and counted in a Packard Tri-Carb Model 3203 Spectrometer after temperature equilibrium had been reached. Under these conditions, the determined value for ^{89}Sr was 95% of actual and that for ^{134}Cs was 89%.

The liquid samples were counted using 1 ml of the aqueous material, 10 ml of 2-methoxyethanol, and 10 ml of a scintillation solution in toluene of twice the concentration described above.

Results

The results found for external ^{89}Sr content for vegetables are tabulated in Table 1 and similar results for ^{134}Cs in Table 2. The cumulative value for the combination operation is based on a direct measurement following the individual steps indicated in paren-

Table 1 Percent Changes in ^{89}Sr Content of Vegetables After Various Food-Processing Steps

Method No.	1	2	3	4	5	6	7	8	
Operation	Dry cleaned	Water wash	Detergent wash	Direct vining	Froth flotation	Brine grading	Steam blanch	Water blanch	Combination
External Content									
Spinach	−27	−79	−82	–		–	−27	−56	1 + 3 + 8 = −92
Peas	–	–	–	140	−61	−61	–	−68	4 + 5 + 6 + 8 = −70
Broccoli	–	−93	−94	–	–	–	−66	−72	8 + (2 or 3) = −92
Internal Content									
Spinach	−33	−66	−66	–	–	–	−65	−71	1 + (2 or 3) + 8 = −75
Peas	–	–	–	–	−44	–	−43	−67	5 + 8 = −70
Broccoli	–	−68	−72	–	–	–	−70	−51	(2 or 3) + 7 = −75

Table 2 Percent Changes in ^{134}Cs Content of Vegetables After Various Food-Processing Steps

Method No.	1	2	3	4	5	6	7	8	
Operation	Dry cleaned	Water wash	Detergent wash	Direct vining	Froth flotation	Brine grading	Steam blanch	Water blanch	Combination
External Content									
Spinach	−8	−78	−80	–	–	–	−12	−77	1 + 3 + 8 = −95
Peas	–	–	–	151	−61	−64	–	−70	4 + 5 + 6 + 8 = −72
Broccoli	–	−91	−92	–	–	–	−28	−84	8 + (2 or 3) = −92
Internal Content									
Spinach	−25	−44	−37	–	–	–	−44	−50	1 + (2 or 3) + 8 = −60
Peas	–	–	–	–	−5	−22	−3	−40	4 + 5 + 8 = −50
Broccoli	–	–	−21	–	–	–	−89	67	7 + 3 = −90

theses in each table. The effect of normal preparation on the internal content of ^{89}Sr and ^{134}Cs in vegetables is also summarized in Tables 1 and 2.

The results found for external ^{89}Sr and ^{134}Cs content of fruit flesh after various food-preparation operations are summarized in Tables 3 and 4.

Peaches and pears harvested from trees grown in soil treated with compounds of ^{89}Sr and ^{134}Cs had such a low level of radioactivity in their flesh that no useful conclusions could be drawn from a study of the effect of normal preparation operations on the residual radioactivity. These results confirm earlier findings of low strontium content of peach and pear flesh from trees grown in soil heavily treated with strontium nitrate.[3]

Strawberries do incorporate substantial amounts of ^{89}Sr and ^{134}Cs after growth of the plants in treated soil. Typical values found were 1×10^{-10} curie per gram of pulp for ^{89}Sr and 6.5×10^{-11} curie per gram for ^{134}Cs. Rinsing of the strawberries with water had little effect on the internal content (12% reduction) of these radionuclides in the pulp fraction.

Table 3 Percent Changes in ^{89}Sr Content of Fruit Flesh After Various Food-Processing Steps

Method No.	1	2	3	4	5	6	7	
Operation	Water rinsed	Lye peeled, whole	Lye peeled, halved	Hand peeled	Ripened, 68°F	Ripened, 42°F	Detergent wash	Combination
External Content								
Peach	–	−91	−77	–	–	–	–	2 = −91
Pear	−5	−20	–	−20	5	−5	−30	7 + 1 + 2 = −50
Strawberry	−31	–	–	–	–	–	–	1 = −31

Table 4 Percent Changes in ^{134}Cs Content of Fruit Flesh After Various Food-Processing Steps

Method No.	1	2	3	4	5	6	7	
Operation	Water rinsed	Lye peeled, whole	Lye peeled, halved	Hand peeled	Ripened, 68°F	Ripened, 42°F	Detergent wash	Combination
External Content								
Peach	–	−97	–	−97	–	–	–	4 = −97
Pear	–	−50	–	–	3600	3000	0	2 = −50
Strawberry	−36	–	–	–	–	–	–	1 = −36

Discussion

The most important general conclusion which can be drawn from the results reported in this paper is that radioactive contamination is most readily removed from crops if they are harvested and processed (with appropriate changes due to possible transfer of radioactivity) within a few hours after exposure to fallout. External contamination can be readily washed from the surface of most fruits and vegetables if it does not have an opportunity to penetrate into the vascular system of the edible portions of the crops.

Tree fruit should be harvested rapidly (even if in a slightly green condition) in order that radioactivity can be removed efficiently. The decontaminated fruit then could be allowed to ripen before processing.

Consideration should be given to harvesting highly perishable crops in a green condition for decontamination and preservation by freezing, canning, or dehydration. The products of such an unconventional operation would have considerable food value even though their flavor and texture would be unacceptable under normal conditions. (FEM)

References

Reference marked with an asterisk is not available in the Isotopes Information Center.

1. **Federal Register*, Sept. 26, 1961.
2. National Canners Association, Investigations on the Removal of Radioactive Fallout from Vegetables and Fruits During Processing by Normal and by New or Modified Processing Operations, Final Progress Report, June 1, 1964–May 1, 1968, Report D-3017, May 1968.
3. J. W. Ralls, S. Primbsch, T. R. Guckeen, H. J. Maagdenberg, J. Rinehart, F. C. Lamb, and W. A. Mercer, Distribution of Strontium and Calcium in Major Vegetable and Fruit Crops and Criteria for Use of Fallout Contaminated Foods, *Radiol. Health Data Rep.*, 8(7): 355-358 (July 1967).

Neutron Radiography Using Nonreactor Sources*

By J. P. Barton

Abstract: *The state of the art of neutron radiography using sources other than reactors is reviewed. This is a follow-up of an article on neutron radiography in general, printed earlier in* Isotopes and Radiation Technology.[1]

Progress has been made recently in evaluation of small, nonreactor sources for neutron radiography. Several installations are now considering setting up such systems—some 20 groups have had discussions with this writer—and many of these involve important applications. Examples of thermal-neutron radiographs taken by nonreactor source techniques have been published in four papers during this decade.[1–4] Discussion of nonreactor sources in these papers has centered on Sb–Be, with only brief mention of alternative small sources. The present paper has the objective of helping to put the current position into perspective, and it is hoped that such an introduction will be of particular use to groups now considering embarking in this field.

It was demonstrated as long ago as 1956 that useful-quality neutron radiography could be performed using a nuclear reactor as the source of neutrons,[5] and within the last few years the technique has been applied successfully in a number of fields.[6] If neutron radiography could be performed with small neutron sources of cost and convenience comparable with those of the standard X-ray generators, the technique might, hopefully, become as widely useful as X radiography is today. Many different types of small neutron sources are now available, both neutron generators and radioactive sources, and further advances will no doubt be made in the near future.[7,8]

Small-Source Thermal-Neutron Radiography: Early Work

Investigations into the possibilities of performing thermal-neutron radiography with nonreactor sources

*These studies of nonreactor-source neutron radiography have been carried out with the kind cooperation of the University of Birmingham (England), Section of Applied Nuclear Science; UKAEA, Harwell and Aldermaston; Centre d'Etudes Nucléaires de Grenoble, Section d'Application des Radioéléments and Service des Accelerateurs; Argonne National Laboratory, Metallurgy Division. The review has also been printed in *Neutron Radiography Newsletter*, No. 8: 7-13 (April 1968). Dr. Barton is currently at Argonne National Laboratory.

have been undertaken by a number of people. Kallmann[9] and Peter[10] experimented with accelerator sources of outputs of about 10^7 and 10^{11} n/sec, respectively. Their reports did not include details about the collimation achieved or the quality of the neutron beam but mentioned difficulties due to the high proportion of interfering radiation—X rays, gamma rays, fast neutrons, and partially slowed neutrons. Early experiments were made by Watts[11] using 1 curie of Pu–Be; Berger[12] using 0.5 curie of Ra–Be and a Van de Graaff accelerator for the ^{7}Li(p,n) reaction; Barton[13] and Dunnicliffe[14] using a D–T and D–D accelerator and a 1-curie Po–Be source; and Rathmann[15] using a linear accelerator with beryllium target. In all cases results were essentially negative since the neutron beams were not sufficiently well collimated to furnish genuine thermal-neutron radiographs of realistic objects (thicker than 1 cm). All these writers stressed the effects of interfering radiation as a serious limitation.

One writer, Warman,[2] did, however, report enthusiastically about neutron-radiography experiments using a 1000-curie Sb–Be source. For this particular $[(\gamma,n)]$ source, the gamma radiation is particularly intense and very penetrating. It was therefore necessary to use the gamma-insensitive activation-transfer photographic technique.* This technique has a low efficiency, i.e., it wastes a large proportion of the available neutrons, and it cannot integrate over long exposure times. The collimator was a cadmium tube with a 2-in. diameter and a length of only 3 in. Because of the high gamma yield, which would necessitate considerable shielding, a source powerful enough for transfer radiography would not be easily portable. There may, however, be applications for which Sb–Be sources are suited, such as radiographic examination of highly radioactive objects where remote-handling techniques and activation-transfer photography are already necessary.

Fast-Neutron Radiography

Small sources appear particularly well suited for fast-neutron radiography. Since moderation and collimation are not required, much more efficient use of the available neutrons is possible. Criscuolo and Polansky reported[16] some experiments on fast-neutron radiography using a D–T neutron generator, and more recently other workers[17–19] have published reports in this field. Some experiments on fast-neutron radiography have also been undertaken by Barton at Birmingham (England), using a D–T and D–D neutron generator, and by Berger at Argonne, using an Am–Cm–Be source. These two workers have been unable to establish that their radiographs were made with pure fast neutrons rather than with mixtures of partially slowed neutrons, X rays, and gamma rays.

Research into methods of fast-neutron detection for radiography is now planned by at least two persons: H. Berger, now at Grenoble, and W. H. Wilkie at Georgia Institute of Technology. If fast-neutron radiography does prove to be practical and to have important applications, one radioactive neutron source that would seem to have particularly promising potential[20] is ^{252}Cf. For example, where a 10^{10}-n/sec radioactive source of another kind may have a diameter of 10 mm, a ^{252}Cf source of 10^8 n/sec might have a diameter of less than 1 mm but be capable of superior radiography performance in fast-neutron point-source geometry.

Thermal-Neutron Radiography: Recent Developments

Activation Transfer

Neutron Generators. The possibility of using a 10^{11}-n/sec neutron generator for the activation-transfer method of thermal-neutron radiography, suitable for highly radioactive objects, has been demonstrated.[21] Dysprosium foil 100 μ thick was used with saturation exposures of a few hours; the collimation ratio was 10:100 (input diameter to length).

Radioactive Sources. Cutforth at ANL (Idaho)[6a] and Farny at Saclay[6b] are experimenting with Sb–Be as the neutron source. Cutforth initially used about 400 curies of ^{124}Sb and has just installed a 6-thousand-curie source. Farny has worked with 10 thousand curies of ^{124}Sb and is considering a source of 100 thousand curies. Whether or not such sources will be practicable and economic will depend on the particular situation (availability of Sb irradiation facilities, for example).

*Since neutrons themselves do not affect a photographic plate (see Ref. 1), neutron radiographs cannot be made directly. Instead, the images are first formed on a suitable neutron-converter screen held adjacent to a photographic film (the "direct-exposure" method) or on a screen that becomes activated and is later placed adjacent to a photographic film (the "activation-transfer" method).

Recent experiments by Barton and Klozar at Argonne have demonstrated that activation-transfer pictures of low density are possible using the 10^9-n/sec ^{241}Am–^{242}Cm–Be source with a 10:100 cadmium collimator.

Direct Exposure

Neutron Generators. Thermal-neutron-radiography characteristics have been evaluated[22] for a range of collimator designs, with experiments on different variables (moderator, geometry, shielding, and detector) for three different neutron generators—a small one with an output of 10^9 n/sec and two larger ones with outputs of 10^{10} to 10^{11} n/sec. The evaluation shows that useful-quality neutron radiography is practical with these generators. Collimations as high as 3:100, which is equivalent to that of the ANL Juggernaut reactor beam,[23] for example, have been achieved. Several other workers, in addition to this writer, have been investigating, or are planning to investigate, the use of neutron generators for direct-exposure thermal-neutron radiography (e.g., Refs. 6*c-e*, 24-27), but as yet no reports have been distributed.

Radioactive Sources. The first known successful direct-exposure neutron radiography with a radioactive source[6*f*,28] used ^{241}Am–Be, a neutron source that had only recently become available. Thermal-neutron-radiography characteristics were established using a 3-curie source (7×10^6 n/sec) for collimator ratios over the range 12:100 to 3:100. This source has a low-energy gamma emission, which is an important advantage for direct-exposure neutron radiography. The low-energy gamma emission also allows the source to be made relatively portable. Americium-241 is a by-product of plutonium burnup in power reactors and should become increasingly available in the future. A neutron-radiography facility using 30 curies of ^{241}Am–Be is now installed at Saclay.[6*g*]

The neutron output of a ^{241}Am–Be source may be increased by a factor of about 100 by irradiation in a reactor neutron flux.[29] The effective half-life of the resulting ^{241}Am–^{242}Cm–Be source is only 163 days, and the original ^{241}Am–Be (half-life 470 years) is largely destroyed. However, in some circumstances there may be a definite demand for this high output over a short time. Experiments to evaluate ^{241}Am–^{242}Cm–Be for neutron radiography have been undertaken at Saclay[30] and at Argonne.[31] The experimenters at Argonne are using Am–Cm–Be sources of strengths 10^7 and 1.1×10^9 n/sec and moderators of BeO and H_2O. It has been demonstrated that good-quality direct-exposure neutron radiography is, in fact, possible with this type of source.

Although ^{241}Am–Be and ^{241}Am–^{242}Cm–Be are the only sources that, to our knowledge, have so far been demonstrated as capable of thermal-neutron radiography, it seems probable that other sources, e.g., ^{210}Po–Be and ^{239}Pu–Be, would be suitable also. A particularly promising source that may become relatively inexpensive[8] in the future is ^{252}Cf. This material is being produced at a rate of tens of milligrams per year. Argonne expects soon to have a source of this nuclide prepared by the Savannah River Laboratory and will undertake experiments to evaluate its neutron-radiography characteristics. Seaborg[20*b*] predicts that about 3 g will be available by 1975.

Conclusions

These conclusions refer exclusively to thermal-neutron radiography of ordinary objects (i.e., not highly radioactive objects). It is possible to summarize here only very briefly and by drawing only on the experimental results and views of the author.

The potential of small-source neutron radiography is seen to have two basic limitations: besides the interfering radiation already recognized, there is the shortage of neutrons. Both problems increase as the degree of beam collimation is increased. In order to satisfy the requirements of a given application, the following variables must be optimized, namely: (*a*) type of source, (*b*) intensity of source, (*c*) type and configuration of moderator, (*d*) beam extraction technique, (*e*) collimation system, (*f*) filter system, (*g*) shielding system, and (*h*) detection system.

The shortage of neutrons, which sets a statistical limit on the information transmitted, is considered (by this author) to be the more fundamental limitation. The approach has therefore been to make maximum use of the available neutrons by (1) using direct-exposure techniques (variable *h* above), thereby enabling long integration times to compensate for low neutron intensities; (2) using the most efficient neutron-converter system (a report on optimized scintillator converters is available[32] and further work is under way); and (3) overcoming the interfering radiation (the second limitation) by manipulation of the other seven variables (*a-g*) rather than by a compromise on the choice of the most efficient detection system. Reports on moderator[33,34] and collimator[35] systems are available, and other reports are in preparation.

Not all applications for neutron radiography will require the highest possible picture quality. Those which do will probably best be solved by use of a nuclear-reactor neutron source. Moderate-quality radiography can be achieved with small sources (Table 1), and, if long exposure times are acceptable (e.g., overnight), very small, inexpensive sources may be the most suitable type of apparatus for the condition of that particular application. (MG)

Editor's Note: A small aluminum-encapsulated source of approximately 1 mg of ^{252}Cf has recently been fabricated at ORNL.[36] With a polyethylene moderator, the thermal-neutron flux is calculated to be

Table 1 Radiography Characteristics of Small Neutron Sources

Source strength yield, fast neutrons per 4π solid angle	Sources evaluated	Thermal-neutron-flux collimator input, n/(cm^2)(sec)	Typical collimation ratio, input diameter to length	Beam intensity at collimator output, n/(cm^2)(sec)	Typical exposure
10^7 n/sec	^{241}Am–Be	10^4	3:100	1	3 days, scintillator + fast film
			10:100	10	9 hr, scintillator + fast film
10^9 n/sec	Small neutron generator (SAMES B.S.1), ^{241}Am–^{242}Cm–Be	10^6	3:100	10^2	1 hr, scintillator + fast film
			10:100	10^3	6 min, scintillator + fast film
10^{11} n/sec	D–T neutron generator (SAMES)	10^8	3:100	10^4	0.5 min, scintillator + fast film
			10:100	10^5	2 min, scintillator + fine-grain film
	Reactor at 1 kw (Juggernaut, Argonne)	10^9	3:100*	10^5	2 min, scintillator + fine-grain film
	Reactor at 200 kw (Juggernaut, Argonne)	2×10^{11}	3:100*	2×10^7	1 min, direct-exposure Gd foil + fine-grain film (0.0005 in.) (Kodak Single R)
	High-quality reactor facility (Melusine 4 Mw or Siloe 15 Mw, Grenoble)	10^{13}	1:100	10^8	Sufficient intensity and collimation to penetrate through thick object and still use thin activation-transfer foils and very-fine-grain film

*The Juggernaut beam collimator is a simple parallel-sided hole penetrating the reactor shield. It is not cadmium lined. All other collimators quoted are lined with a material opaque to thermal neutrons (Cd or B) and are of the divergent type.

used. Such sources may be fairly portable and easy to manipulate.

The cost of a neutron source rises rapidly with the neutron yield, and it will be important to design the facility to suit the particular application and circumstances. For example, the geometric unsharpness, the inherent foil and film unsharpness, and the neutron statistical unsharpness should be carefully matched to each other and to the inspection task requirements.

Table 1 outlines some of our experimental results. By reference to survey experience such as this, it should be possible to decide (1) whether or not small-source techniques can satisfy the specified requirements of a particular application and (2) what will 0.02 n/(cm^2)(sec) for each n/sec from spontaneous fission. A state-of-the-art review on the uses of neutron radiography, a publication of the Isotopes Information Center, is in press.[37] Some additional references that will be of interest to the reader are:

W. C. Reinig, Advantages and Applications of ^{252}Cf as a Neutron Source, *Nucl. Appl.*, **5**: 24-25 (1968).

W. C. Reinig, ^{252}Cf: A New Isotopic Source for Neutron Radiography, paper to be presented at the National Conference of the American Society for Nondestructive Testing, Detroit, Oct. 14–17, 1968.

C. S. Schlea and D. H. Stoddard, Californium Isotopes Proposed for Intracavity and Interstitial Radia-

tion Therapy with Neutrons, *Nature* **206**: 1058-1059 (1965).

C. N. Wright, A. R. Boulogne, W. C. Reinig, and A. G. Evans, Implantable ^{252}Cf Sources for Radiotherapy, *Radiology*, **89**: 337-339 (1967).

References

References marked with an asterisk are not available in the Isotopes Information Center.

1. J. P. Barton and J. L. Boutaine, Initial Development of Neutron Radiography in France, *Isotop. Radiat. Technol.*, 5(3): 214-218 (Spring 1968) [in French in *Suppl. Bull. Inform. A.T.E.N.*, No. 66, pp. 4-9 (July–August 1967)].
2. E. A. Warman, Neutron Radiography in Field Use, *Mater. Eval.*, **23**: 543-551 (1965).
3. J. P. Barton, Toward Neutron Radiography of Radioactive Objects in Hot Cells, *Trans. Amer. Nucl. Soc.*, **10**: 443 (1967).
4. D. C. Cutforth, On Optimizing an Sb–Be Source for Neutron Radiographic Applications, *Mater. Eval.*, **26**: 49-53 (1968).
5. J. Thewlis, Neutron Radiography, *Brit. J. Appl. Phys.*, **7**: 345-350 (1956).
6. *Neutron Radiography Newsletter*, Nos. 1–8 (1964–1968), American Society for Nondestructive Testing, Evanston, Ill. (*a*) D. C. Cutforth, Nos. 1–8; (*b*) M. Farny, No. 7, p. 3; (*c*) J. H. Hamilton, No. 4, p. 4; (*d*) G. Lachese, No. 6, p. 2; (*e*) M. Mouraille, No. 7, p. 3; (*f*) J. P. Barton, No. 6, p. 3; (*g*) J. L. Boutaine, No. 6, p. 3.
7. Accelerator Targets Designed for the Production of Neutrons, Proceedings of the Meeting Held at Grenoble, France, June 21–22, 1965, Euratom Report EUR-2641.d, f, e (Conf-650635), 1966.
8. R. L. Stetson, Transplutonics: Promising Neutron Sources for Research, *Nucleonics*, **24**(11): 44-47, 75 (1966).
9. H. Kallmann, Neutron Radiography, *Research*, **1**: 254-260 (1948).
10. O. Peter, Neutron Radiography (in German), *Naturforsch.*, **1**: 557-559 (1946).
11. H. V. Watts, Research on Neutron Interactions in Matter as Related to Image Formation, Report ARF-1164-27, Armour Research Foundation, August 1962.
12. H. Berger, Neutron Radiography: A Second Progress Report, in Second Symposium on Physics and Nondestructive Testing, Held at Argonne National Laboratory, October 3–5, 1961, USAEC Report ANL-6515, pp. 28-52, Argonne National Laboratory.
13. J. P. Barton, Concepts in Neutron Radiography, University of Birmingham (England), 1963, unpublished.
14. C. J. Dunnicliffe, Development of a Portable Neutron Source Suitable for Use in Radiography, University of Birmingham (England), August 1964, unpublished.
15. D. W. Rathmann, Preliminary Experiments in Neutron Radiography with a Radiographic Linear Accelerator, Thiokol Chemical Corp., August 1964, unpublished.
16. E. L. Criscuolo and D. Polansky, Fast Neutron Radiography, Missiles and Rockets Symposium, U. S. Naval Ammunition Depot, Concord, Calif., Apr. 18, 1961.
17. J. Anderson, S. B. Osborn, R. W. S. Tomlinson, and J. W. Smith, Neutron Radiography in Man, *Brit. J. Radiol.*, **37**: 957-958 (1964).
18. E. Tochilin, Photographic Detection of Fast Neutrons: Application to Neutron Radiography, *Phys. Med. Biol.*, **10**: 477-490 (1965).
19. D. E. Wood, Fast Neutron Radiography with a Neutron Generator, *Trans. Amer. Nucl. Soc.*, **10**: 443 (1967).
20. *a.* J. P. Barton, Transplutonium Isotopic Neutron Sources: Foreseeable Future Demand for Neutron Radiography, April 1968, unpublished.
    b. G. T. Seaborg, Mass Production and Practical Applications of Actinide Elements, *Isotop. Radiat. Technol.*, **6**: 1-16 (1968).
21. *J. P. Barton, On the Possibility of Using a D–T Neutron Generator for the Radiographic Inspection of Highly Radioactive Elements, SAR-G/66-370, Nov. 1966.
22. J. P. Barton, Neutron Radiography Using Small Neutron Sources, Centre d'Etudes Nucléaires, Grenoble (report in preparation).
23. H. Berger, Characteristics of a Thermal Neutron Beam for Neutron Radiography, *Int. J. Appl. Radiat. Isotop.*, **15**: 407-414 (1964).
24. L. Holland, University of Birmingham (England), personal communication, 1967.
25. S. Kawasaki, Tokyo Shibaura Electric Co., Kawasaki, Japan, personal communication, 1967.
26. M. Radwan, Institute of Nuclear Research, Warsaw, Poland, personal communication, 1968.
27. L. Vu Hong, Centre d'Etudes Nucléaires, Grenoble, personal communication, 1967.
28. *Neutron Radiography: Possibilities and Perspectives* (in French), Proceedings of Information Meeting, Saclay, Oct. 5, 1966, French CEA Report DR/AR/67.1.
29. D. C. Stewart, R. W. Anderson, and J. Milsted, The Production of Curium by Neutron Irradiation of ^{241}Am, USAEC Report ANL-6933, Argonne National Laboratory, 1964.
30. J. L. Boutaine, Centre d'Etudes Nucléaires, Saclay, personal communication, 1967.
31. J. P. Barton and M. Klozar, Argonne National Laboratory, unpublished.
32. *J. P. Barton, Scintillators in Neutron Radiography, DR/SAR G/67-46, December 1967.
33. *M. Mouraille, Thermalization of Fast Neutrons Produced by a Low-energy Electrostatic Accelerator (in French), ACC/67-06, July 1967.
34. *J. P. Barton, Moderation of Neutrons from Point Sources—Application: Neutron Radiography, DR/SAR-G/67-45, December 1967.
35. J. P. Barton, Divergent Beam Collimator for Neutron Radiography, *Mater. Eval.*, **25**: 45A (1967).
36. J. P. Nichols, Design Data for ^{252}Cf Neutron Source Experiments, *Nucl. Appl.*, **4**: 382-387 (1968).
37. Martha Gerrard, Uses of Neutron Radiography, USAEC Report ORNL-IIC-16, Oak Ridge National Laboratory, 1968.

Section

III

Isotopes and Radiation Technology

Food Irradiation

Designing for Lower Food-Irradiation Costs

By John B. Huff*

Abstract: *A comparison of data on ^{60}Co use in the Marine Products Development Irradiator shows that using an 8-ft source instead of a 4-ft source will result in lower costs. Using ^{137}Cs instead of ^{60}Co may reduce costs, depending on price changes. A technique is described that uses a point source to simulate an annular source. A simple compact irradiator for shellfish is described; cost data show that irradiation will cost about 1.6 cents/lb under typical conditions.*

The use of ionizing radiation to preserve various food products has been investigated for a number of years, and laboratory studies have shown that seafood is one of the more promising food groups for irradiation preservation. Through a joint effort by the Atomic Energy Commission and the Bureau of Commercial Fisheries, a pilot-plant scale facility for processing seafoods became operational in April 1965. This facility, the Marine Products Development Irradiator (MPDI), is located in Gloucester, Mass., and has been described in detail by Miller and Herbert.[1] Its plane source was initially loaded with 250 kilocuries of ^{60}Co.

When in use, the source is sandwiched horizontally between two absorbing slabs of prepackaged fishery products. These slabs are 110 in. long, 22 in. wide, and 6.5 in. thick, and each one is actually made up of 14 packages. These packages are mechanically moved from one package position to another so that each package occupies all 28 positions, in turn, for a predetermined dwell time. This procedure is followed to obtain uniform dose distribution. By adjusting dwell time, the dose can be increased or decreased. The source is stored under water in a vertical position when not in use, but a curved track runs the plaque to a horizontal position between the product slabs. Six subframes make up the source plaque, which is 48 in. long and has an effective width of 11 in. The position of the source is symmetrical with respect to the product slabs; they overlap the source 5.5 in. on each side and about 31 in. on the ends. With this configuration, about 22% of the gamma energy is absorbed by the product.

The purpose of the MPDI is to develop and demonstrate seafood irradiation techniques,[1] so that private investors will be encouraged to enter the commercial food-irradiation field. One of the main factors determining whether or not investors will risk investment is capital cost. An important factor in cost, in turn, is the design of the irradiator. The purpose of this article is to suggest design changes that will reduce these costs.

Unfortunately, operation of the facility at capacity for more than a few hours at a time has not been possible. Because the Food and Drug Administration has not yet approved irradiated seafoods for human consumption, treated seafoods have to be disposed of at present; so, extended runs are very expensive. The cost data presented here are, therefore, based largely on a part-time operation and, accordingly, are subject to revision.

In developing an irradiator design to help minimize cost, the following main topics were considered: (1) analysis of source procurement needs, which vary with source plaque design and the useful life of the facility; (2) comparison of procurement costs for ^{60}Co and ^{137}Cs on an energy-yield basis; (3) use of a point source to generate a geometric shape for making dose-distribution measurements; (4) use of data obtained from dose-distribution measurements to design a proposed irradiator that is simple and compact; and (5) projection of available figures to show a cost-range estimate for food irradiation.

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Source Procurement Needs

Commercial use of a gamma irradiator requires periodic source replenishment. If the MPDI source were kept through one isotope half-life, which is about 5.25 years for ^{60}Co, the plant would then have only half its original capacity. Processing costs would therefore be almost doubled, as operating expenses would remain substantially unchanged.

Procedure with Four-Foot Source Plaque

The MPDI Preliminary Design Report[2] included a schedule for source replenishment. Under this scheme, 96 source strips would be separated into three groups of low, medium, and high specific activities. For example, one pair of subframes might contain 30 kilocuries each, the second pair 40 kilocuries each, and the third pair 55 kilocuries each, making a total of 250 kilocuries for six subframes used in the assembled plaque. At the end of the first year, one of the 30-kilocurie subframes would be replaced with a new one having about 55 kilocuries of activity. Each following year the subframe having the lowest curie content would be replaced by a new one having sufficient activity to bring the source strength up to 250 kilocuries. Subframes used for annual replenishment would contain from 53 to 59 kilocuries. Since each subframe contains something less than 1 kg of cobalt, a specific activity of some 55 to 60 curies/g is required for replenishment. Under this replenishment program, source strength would vary from 220 to 250 kilocuries, and the plant should always operate above 88% of design capacity.

It is evident that the annual replenishment requirement at MPDI is the sum of activity lost by decay plus that removed to make space for the new subframe. Replenishment requirements for MPDI under the conditions given above have been calculated for a 20-year period (Fig. 1). For 10 years' operation, 750 kilocuries are needed and 230 would be discarded. Over a 20-year period, 1300 kilocuries would have to be purchased and 475 kilocuries discarded. From these figures it can be seen that about one-third of the ^{60}Co purchased over a 20-year period is discarded when using this system of source replenishment at MPDI.

Larger Capacity Source Plaque

If a source plaque were designed to have space for adding new subframes, source material could be used longer and more of its energy could be utilized. Accordingly, source requirements were calculated for a

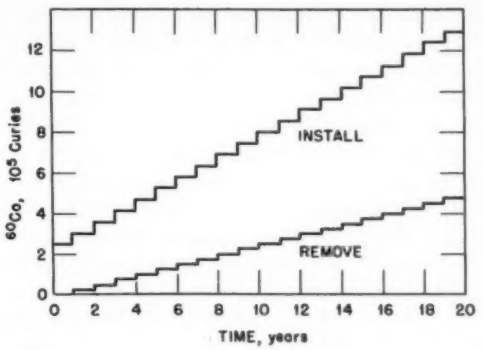

Fig. 1 Annual changes required in the MPDI source to maintain its capability. Initial loading is 250 kilocuries.

hypothetical irradiator comparable to the MPDI except that the source length was extended from 4 ft to 8 ft. Six subframes would then be loaded initially, and a new one added each year until the seventh, when it would become necessary to replace a subframe. Cobalt-60 requirements for maintaining this source between 220 and 250 kilocuries are shown in Fig. 2. Six

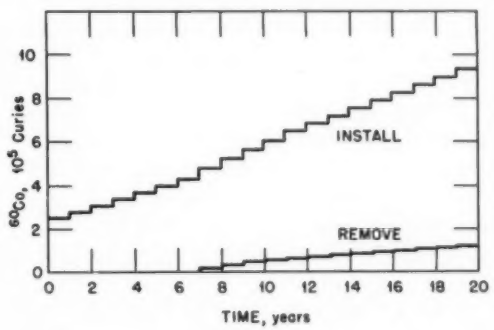

Fig. 2 Annual changes required in a hypothetical MPDI source 8 ft wide. Initial loading is 250 kilocuries.

subframes, each containing 42 kilocuries, would be required for the initial loading; annual replenishment would require subframes containing 30 to 47 kilocuries; the required specific activity is considerably lower than that for the MPDI. After the thirteenth year, replenishment needs per year become constant at 36 kilocuries.

A comparison of Figs. 1 and 2 shows that doubling the length of the MPDI source frame would save 185 kilocuries over the first 10 years of operation. The

MPDI source-storage pool is 9 ft in diameter; so, modification of the facility to use an 8-ft source may be feasible. An alternative method for adding source material should be considered. The present MPDI source frame may be modified to double the thickness of the plaque so that six additional subframes could be added. This idea, however, is more realistically applicable to the design of future facilities. There are other ways to lower costs of source replenishment, of course. Biannual replenishment would involve slightly different economics. A lease-type agreement with a commercial ^{60}Co supplier whereby the user pays only for curies used (plus a service charge) would also change the economics. However, such schemes fall outside the scope of design.

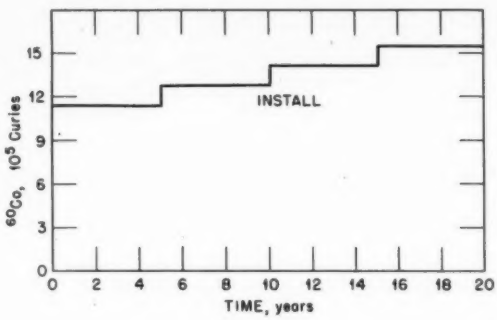

Fig. 3 Annual changes required in a hypothetical MPDI source using ^{137}Cs and having a gamma energy yield comparable to that of the MPDI. (Note change of scale along vertical axis.)

Comparative Requirements for ^{137}Cs

Upon decay, ^{137}Cs yields 0.662-Mev gamma rays from 85% of its disintegrations.[3] Cobalt-60 yields two gamma rays for each disintegration, one having an energy of 1.17 Mev, the other an energy of 1.33 Mev. The average gamma-energy yield for ^{137}Cs is 0.562 Mev per disintegration compared with 2.5 Mev per disintegration for ^{60}Co. Thus 4.44 curies of ^{137}Cs have an immediate energy yield equivalent to 1 curie of ^{60}Co. However, the curie unit indicates rate of decay rather than potential energy. A curie of ^{137}Cs represents 4.47×10^{19} radioactive atoms, whereas a curie of ^{60}Co is associated with 8.8×10^{18} radioactive atoms. Therefore the lifetime yield in ergs is about 12% greater for a curie of ^{137}Cs than for a curie of ^{60}Co.

Over the life of a processing facility, the cesium isotope's higher potential energy will be reflected in the procurement cost of the source. Accordingly, source requirements for a ^{137}Cs facility having the same energy yield as the MPDI were calculated (Fig. 3).

There is a difference between energy yield and absorbed dose. The percentage of total emission absorbed by the product at the specified minimum dose is normally called *utilization efficiency*,[1] and it varies with irradiator design and other factors. Consequently, efficiency and certain other factors have been disregarded in this comparison of ^{60}Co and ^{137}Cs.

Cesium-137 requires a large initial investment followed by modest replenishment costs. Since replenishment to maintain at least 88% of capacity can be made at 5-year intervals, there should be a corresponding reduction in the number of radioactive shipments and other expenses associated with source transfers. On the other hand, a cost engineer would probably add projected interest charges (which are higher for cesium) to procurement costs when estimating the total investment in the source over a 10- or 20-year period. Disposal of spent sources, shipping cost, premiums paid for high specific activity, interest, shielding requirements, efficiency, density of source material, and many other factors should be considered when a facility is designed for minimum processing costs. These factors are usually variable; so they were disregarded in the preparation of the graphs shown. Figures 2 and 3 are thus compared on an energy-yield basis; the ratio of ^{137}Cs to ^{60}Co, in curies to be purchased, decreases with time from 4.44 initially to 1.66 for a 20-year period. Under the conditions given, ^{60}Co would be competitively priced at 4.44 times the ^{137}Cs price for 1 year of use or at 1.66 times the cesium price over a 20-year period. In this comparison the initial loading of ^{60}Co uses 50% of available source space whereas the ^{137}Cs uses about 74%. If ^{60}Co costs 50 cents/curie, then ^{137}Cs should be priced at 50/4.44 or about 11 cents/curie when compared for a 1-year period. On the other hand, extending the period to 20 years would make ^{137}Cs competitive at 50/1.66 or 30 cents/curie. This comparison assumes equal utilization of the energy output from the two sources.

Sources developed for using ^{137}Cs salts may not resemble MPDI source strips but will probably be more massive than those using metallic cobalt. More development work has been done on ^{60}Co than on ^{137}Cs source arrays. Cesium will probably be used as cesium chloride powder, or pellets, having a specific activity of about 25 curies/g. The source material must be suitably encapsulated to prevent leakage. In the cylindrical source application described below, a

cesium salt might fill the space between two concentric pipes having ring-shaped plugs at the ends. Uniform point-to-point gamma intensity would be expected from such a design.

Dose-Distribution Measurements

Irradiator design involves a series of compromises. A source having high specific activity is obviously desirable because self-absorption, encapsulation costs, and size of shielding enclosure tend to increase as the source becomes more bulky. Although starting with a small source of high specific activity is relatively easy, maintaining it is expensive, because partially decayed source strips must be replaced to keep the source small. A cylindrical shell source array should require less shielding than a plane source of the same size. An experiment was therefore designed to measure the radiation fields in the vicinity of such a source.

A 10-mc ^{60}Co calibration source was used to generate the cylindrical shell. The mechanism used may be visualized as follows: A drill press is adjusted to have a spindle speed of 5 rpm. The hub of a wheel 10 in. in diameter is secured in the chuck of the drill press. A 10-mc source is suspended, like a plumb bob, from a hook on the rim of the wheel. Raising or lowering the rotating drill press spindle at a uniform rate makes the source describe a helical path around the dosimeter, which appears as a cylinder when seen by a dosimeter in a fixed position. In actual practice a small motor was used to rotate the suspended source; the motor itself was suspended by a steel measuring tape running over a pulley. This arrangement permitted the experimenter to work at a safe distance from the source while adjusting the vertical movement.

Raw clam meats are frequently packed in 1-gal tins that are 6.5 in. in diameter and 7.5 in. tall. Three tins were stacked to make an experimental column. The tins were filled with raw seafood, water, or rice. Bulk rice has a specific gravity of about 0.8; it makes a convenient phantom, as gamma personnel dosimeters can be embedded at the desired positions. When water or wet seafood was used, the dosimeters were placed in corked test tubes and taped to small wooden strips, as required, to keep them in position. A cylindrical source was then generated around the product column, and the dose was determined for various positions.

In the first experiment a source shell was generated, 8.5 in. in diameter by 12 in. high, so that it would have a common geometric center with the second of three cans. With the source lifted 1 in./min for 12 min, the center of the can showed a dose of 121 mr, and the outside showed a dose of 162 mr, which gives a maximum/minimum ratio of 1.34. Expanding the source shell to 10.25 in. diameter in the second experiment resulted in a center reading of 88 mr and an outside reading of 106 mr, or a maximum/minimum ratio of 1.2. It should be noted that the absorbed dose at the center dropped some 27% when the diameter of the source shell was increased to 10.25 in.; the angle of shadow from the moving source at any instant is obviously greater when the absorber is nearer the source. Most source arrays can be thought of as being a large number of point sources.

These findings indicate that an acceptable dose distribution can be obtained in a single pass of gallon cans through an annular ^{60}Co source. A circular array of MPDI source strips is considered for comparison. Source strips must be matched in specific activity, however, to approach the uniform circumferential intensity obtained with the rotating test source.

If 32 MPDI source strips were placed edge-to-edge, like the staves of a barrel, they would form an almost circular polyhedron, about 8.5 in. in diameter, having a cobalt strip length of 11 in. The dimensions would thus be comparable with the 8.5-in.-diameter generated source described above. Since 32 strips represent one-third of MPDI source strength, or 83.3 kilocuries, and since the generating source is 0.01 curie, intensity from the point source should be increased by a factor of 8.33×10^6. In the experiment a minimum field of 121 mr in 12 min was recorded; this is equivalent to 605 mr/hr, which, if multiplied by the factor 8.33×10^6, should give a field of some 5×10^6 r/hr at the geometric center of the source. The field of 5×10^6 r/hr intensity can be converted to 4.8×10^6 rads/hr (i.e., 4.8 Mrads/hr) by using the factor 0.96 for the absorption of ^{60}Co gamma rays in soft tissue.[4]

Axis dose-rate readings are higher at the center of a generated cylinder source than at the ends. But material moving along the axis at a uniform rate is exposed as it approaches and leaves the annular source. Assuming a symmetrical system and a uniform product flow along the axis, the dose accumulated by a particle as it approaches and leaves the annular source tends to compensate for the weaker fields within the ends of the cylindrical source shell. From dose measurements taken along the axis, it appears that total absorbed dose is approximately equivalent to the midway dose rate multiplied by product volume contained between the end planes of the source cylinder. About 13 lb of product is contained within the generated source used in the experiment; the use of 83.3 kilocuries should thus give a capacity of 4.8×13 or 62.4 Mrad-lb/hr. If

all 96 MPDI source strips were arrayed in this manner, a cylindrical source 8.5 in. in diameter by about 3 ft in length having a capability for irradiating 187 Mrad-lb/hr should result. This capability is equivalent to irradiating 748 lb/hr at a dose of 0.25 Mrad. Although this capability is considerably less than the MPDI capacity of 675 Mrad-lb/hr, it appears that methods can be developed to utilize radiation fields outside the cylinder and that certain advantages would result from a more compact design.

Dose measurements were also taken in water surrounding a generated cylindrical source. A source 6.5 in. in diameter was generated in a submerged vessel 11.5 in. in diameter. Dosimeters were spaced 1.5 in. apart, radially, in the 6 in. of water surrounding the vessel. Maximum/minimum ratios varied from 1.36 to 1.78. In spite of the fact that maximum/minimum ratios are much higher than those within a cylindrical source shell, designers of such facilities should give thought to utilizing this otherwise wasted gamma energy. Packages about 2 in. thick could apparently be processed at a 1.3 maximum/minimum ratio by re-orienting them during a two-pass irradiation. Pouches containing the product might be sealed in continuous strips of plastic film and drawn through the radiation field surrounding a cylindrical source.

In the case of pasteurized foods, the use of a fixed maximum/minimum limiting ratio may not be the approach. If, for example, a minimum dose of 200 krads is specified, the total number of bacteria killed in a package will, of course, be higher with a 400-krad maximum than with a 260-krad maximum. By establishing objectionable taste and odor threshold values and using these to set upper dose limits, the capability of a cylindrical source array—as well as most other arrays—could be extended considerably in cases where the minimum dose is relatively low.

A shellfish facility could thus be used to process gallon cans of clams in the center, while other fishery products packed in pouches or envelopes formed from continuous strips of thermosealing plastic film could be processed on the outside. Certain fishery products might be pumped through a helical tube surrounding the source if sterile tanks could be used for bulk distribution.

Compact Shellfish Irradiator

The foregoing calculations point to the possibility of designing a small, rather inexpensive irradiator (Fig. 4) for use by a shellfish processor having limited volume or for use as a pilot plant by other food processors interested in testing markets and establishing production possibilities.

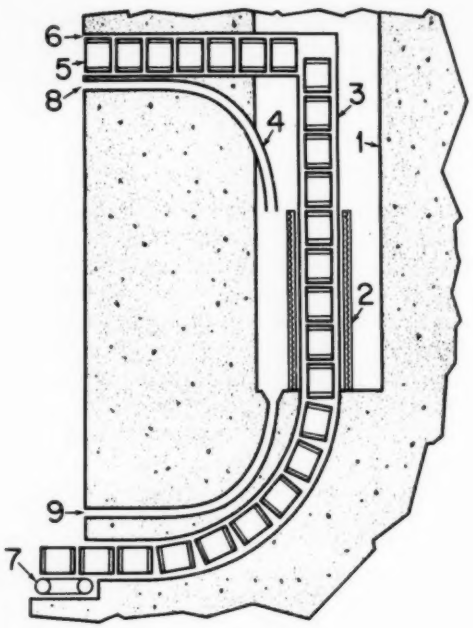

Fig. 4 Vertical schematic of proposed shellfish irradiator. (1) Shaft, shielded by concrete or other material. (2) Stovepipe-shaped source. (3) Product guide for cans. (4) Product guide for contiguous plastic pouches. (5) Product cans. (6) Chute for cans. (7) Metering device. (8) Port for insertion of plastic pouches. (9) Port for output of irradiated plastic pouches of product.

The irradiator is basically a vertical shaft 2 or 3 ft in diameter, surrounded by concrete or other shielding material.

Source Loading

Openings into the shaft are capped so that it can be filled with water while the source is being loaded. A shipping cask (Fig. 5) is unbolted from its shielding base; then the bell-shaped top is set over the shaft by use of a remotely operated crane. A source cylinder (or a basket containing source components) is lowered into the shaft from its position within the bell by a mechanical device on the cask top. With the source shielded by water in the shaft, the bell is hoisted, and its internal tackle is disconnected from the source by use of underwater tools. The operation is repeated as required to assemble or replenish the source. After the source cylinder (which is identified both as item 2 in

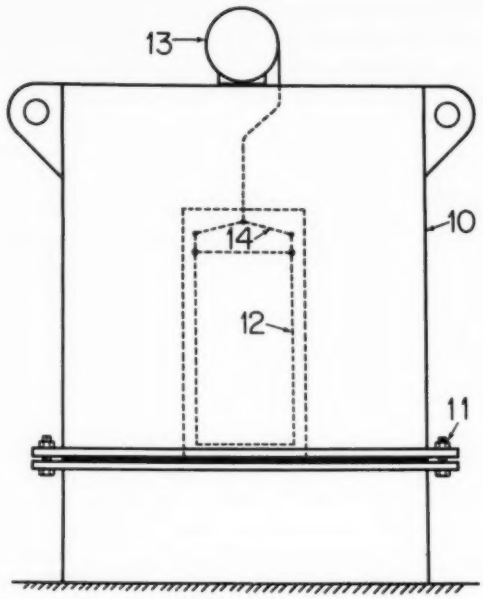

Fig. 5 Shipping cask for transferring source material to irradiator. (10) Bell-shaped cask. (11) Flanged base. (12) Source. (13) Hoist. (14) Supporting cover. In operation, the cask is unbolted from the base and placed over the receiving shaft. Then the source is lowered by the hoist and uncoupled from the supporting cover under water, after which the cover is withdrawn.

Fig. 4 and as item 12 in Fig. 5) is assembled from one or more shipments, product guides can be installed under water and a shielding cap placed over the top of the shaft. The water can then be drained.

Irradiation Procedure for Cans

Gallon cans are fed into the system through a chute which is designed to protect personnel from radiation. The sketch is schematic only, as cannery-machine engineers can best adapt conventional can-handling devices to this system. The cans descend through a guide pipe by gravity, the rate of descent being controlled by a conveyor or some other indexing device at the lower end of the guide pipe. To start the system, the operator fills it with empty cans, using a backflow of air, if necessary, to prevent their banging together.

Products Packaged in Plastic Film

A ball attached to a string and introduced through a port (item 8, Fig. 4) should reappear at the other port (item 9, Fig. 4), so that the string can be used to start a strip of contiguous plastic pouches into one port and out through the other. If the pouches exceed 1 in. in thickness, they need to pass the source a second time with their opposite faces toward the irradiator to maintain a 1.3 maximum/minimum dose ratio. They are drawn through the radiation field at a rate that results in the desired dose. By the extrapolation of data from rotating-source experiments, it appears that the contents of the pouches can be given a dose of about 0.25 Mrad in passing a 250-kilocurie source at a rate of 1 ft/min. The product tubes can approach the source from any direction, so that, although only one set of tubes is shown in Fig. 4, many can be installed.

Disadvantages

Refrigeration and ventilation problems have been disregarded in the design shown in Fig. 4. A rather long conveyor moving at irradiator speed allows considerable warming of a prechilled product; segments of the conveyor and product tube would thus require refrigeration. Similarly, heat generated by the source (equivalent to about 0.015 watt/curie) must be dissipated by a stream of air or by other means. Large sources cause considerable ozone formation in the air surrounding them, so that an exhaust system, including a stack and filters (for particulate matter), is usually required. Fitting this equipment to the shielding so that it will not interfere with loading of the source is more difficult in a compact design.

The 6.5-in.-diameter cans shown in Fig. 4 are approaching the size limit if the maximum/minimum ratio must be kept below 1.3. Converging gamma rays compensate sufficiently for product absorption losses within an annular source to maintain that ratio at the diameter given. However, a 17-in.-diameter source generated around an 11.5-in.-diameter column of rice (5-gal can) showed a maximum/minimum ratio of 1.59; reducing the source diameter to 14 in. gave a maximum/minimum ratio of 1.87. A disadvantage of the proposed shellfish irradiator would thus be its limitation on package size.

Advantages

An irradiator resembling the one sketched in Fig. 4 would apparently fit into a production line, so that little additional labor would be required for irradiation. Since there are no moving parts in the radiation field, health-physics coverage should be required only when the source is being loaded. Electronic devices should

provide adequate health-physics protection between loadings. In fact, it appears that only one employee per shift, in addition to the regular plant crew, would be required to run dosimetry and watch irradiator instruments.

Substantial savings in material handling costs can be realized by using an in-line irradiator. A package must be handled 11 times in passing from a local processor's cold room to the MPDI irradiator and back again. In addition, a refrigerated truck is required, and the driver's time is chargeable to the irradiation process.

Full production on day shift at the MPDI thus requires a supervisor, a health physicist, two operators, a truck driver, and a maintenance man. On evening and night shifts, two operators and a relief man are required, a total of 12 men being required for three shifts. In comparing three employees for an in-line irradiator with 12 for the MPDI, the assumption is made that no extension of the regular plant crew will be required. This assumption is based on the premise that irradiator personnel will contribute effort to other production activities, since their routine duties should not require full-time attention.

The in-line irradiator should shorten the time required to get seafood on the consumer's table. In a clam-shucking plant, for example, gallon cans of clams could go on the irradiator line as soon as they are filled, thus avoiding the delay required to accumulate a truckload. Indeed, if irradiator capacity equals shucking capacity, irradiation involves no significant aging of the product.

A separate facility, such as the MPDI, has considerable overhead expense in addition to labor. Such a facility requires heat, utilities, janitorial service, and other expenses normal for plants of comparable size. Although the addition of an in-line irradiator would add somewhat to the space and power requirements of an existing plant, overhead costs would be less than for a separate facility.

Overall Cost Estimates

Despite the fact that estimated costs for the irradiator shown in Fig. 4 should be based upon an engineering study, such as Ref. 2, some reasonable estimates can be projected from experience with the MPDI. For example, Fig. 1 shows that source procurement required to maintain the MPDI for 20 years is 1.3 million curies, equivalent to 65,000 curies/year. If the cost of encapsulated ^{60}Co is 50 cents/curie, then the projected average source cost for the MPDI becomes $32,500 per year. For the simplification of the estimate (Table 1), average source procurement is shown as an operating expense (like the gas bill, for example) although the derivation of costs is more complex. If the plant, without ^{60}Co, is valued at $505 thousand and is fully depreciated over a 20-year period, annual depreciation costs average $25,200. Supplies and repair parts are estimated at $12 thousand per year. Heat and electricity for the MPDI add another $3460. In a comparable privately owned facility, interest, taxes, insurance, and similar charges might be about 1% per month, or $75,600 per year on a $630 thousand investment (plant plus initial source). Assuming a 5-day work week, three shifts per day, and 12 employees at an average wage of $7500 each, the payroll is $90 thousand. No provision is included here for profit or income taxes on profit. Obviously, the figures vary with plant location and other factors; the reader may therefore adjust them as needed to fit another set of conditions. But a privately owned facility, comparable to the MPDI, could expect operating costs at this level.

Table 1 Major Annual Costs for Operating a Facility Comparable with MPDI

Source procurement	$ 32,500
Plant depreciation	25,200
Supplies and repairs	12,000
Heat and electricity	3,460
Taxes, interest, and insurance	75,600
Payroll	90,000
Total annual cost	$238,760

MPDI Unit Cost Range

The MPDI has a capacity of 675 Mrad-lb/hr, equivalent to 2700 lb/hr at a dose of 0.25 Mrad. Projecting these figures shows that it is theoretically possible, by working fifty 120-hr weeks, to process 16,200,000 lb. Total cost shown in Table 1 divided by this annual production figure gives a processing cost of 1.5 cents/lb, which is considered to be the lower limit for MPDI unit costs.

There has been considerable mechanical difficulty with product conveyors and the source elevator (Fig. 6) at the MPDI (Ref. 5). Careful repair and maintenance planning would be required to run the MPDI for 120 hr each week. One approach would be to schedule maintenance work on weekends so that a major component, such as a conveyor chain, could be

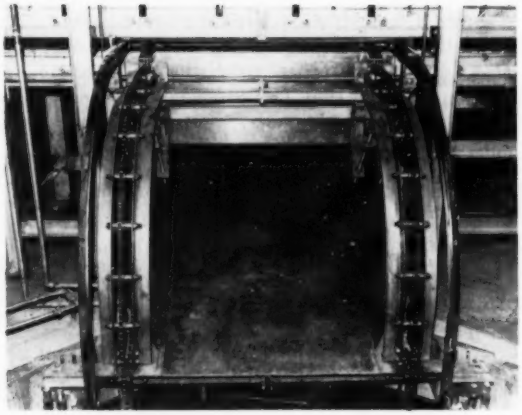

Fig. 6 MPDI source in position between two product slabs. Track goes to underwater storage pool.

removed for overhaul while a spare component kept the plant running. Such a procedure would probably require overtime or other extension of the manpower estimate made in Table 1. In any event, mechanical breakdowns, shortages of raw material, and unforeseen factors tend to raise the minimum unit processing cost of 1.5 cents/lb. On the other hand, if the facility runs only half the time and expenses remain constant, the MPDI unit processing cost becomes 3 cents/lb. This cost is considered a reasonable upper limit.

Using the modified source plaque (Fig. 2) will reduce annual source requirements from 65 to 47 kilocuries, or 18 kilocuries, which is a saving of $9 thousand per year. This saving reduces unit costs by 3.7%. Similarly, if encapsulated ^{137}Cs is available at 12.5 cents/curie, source purchases over 20 years total $195 thousand or an average of $9750 per year (down from $32,500 for ^{60}Co at MPDI), reducing total operating costs by 9.5%. If production and other stated conditions remain constant, changing from ^{60}Co to ^{137}Cs should reduce the minimum unit cost from 1.5 to 1.36 cents/lb.

Shellfish Irradiator Costs

Estimated major costs for the proposed in-line shellfish irradiator are shown in Table 2. Source costs are taken from Fig. 2, which assumes space for doubling the initial source loading. The MPDI has a plane source with an effective area of 429 sq in.; an equivalent area of sheet material rolled into a stovepipe shape would form a right cylinder 9 in. in diameter by 15.2 in. high. The MPDI source is double thickness, with ^{60}Co strips placed back to back. The source holder (Fig. 4) would thus have to be about 10 in. in diameter by 36 in. high, to allow for source holder thickness and the replenishment program given.

The need for expensive equipment has been largely eliminated in the proposed design. The shaft can be made of precast concrete pipe surrounded by earth or concrete shielding. Since it would be filled with water only when the source is being transferred, there is no apparent need for a stainless-steel lining. Also eliminated is the need for a stainless-steel source elevator, refrigerated product storage, multiple-pass conveyor system, and electronic safeguards for unlocking the cell. If installed in a processing plant where space and other conditions are favorable, an in-line irradiator might therefore be constructed for one-fourth the cost of the MPDI. This basis of $126 thousand is used for those cost estimates (Table 2) that are a function of plant cost.

Table 2 Major Annual Costs for Operating a Cylindrical Source Facility, Such as the Compact Shellfish Irradiator

Source procurement	$23,400
Plant depreciation	6,300
Supplies and repairs	3,000
Heat and electricity	865
Taxes, interest, and insurance	18,900
Payroll	22,500
Total annual cost	$74,965

Shellfish Irradiator Unit Costs

The generated-source cylinder experiment described above showed that about 748 lb/hr can be irradiated in gallon cans. If a like amount can be irradiated in plastic pouches outside the source cylinder, capability of the irradiator becomes 1496 lb/hr. Working fifty 120-hr weeks at this rate gives an annual production of 8,980,000 lb, which, when divided by the total cost in Table 2, shows a minimum unit cost of 0.8 cent/lb. If the plant works at only half its capacity, a unit cost of 1.6 cents/lb is predicted for the in-line irradiator.

Summary

Maintaining the MPDI above 88% of its design capability requires purchasing 750 kilocuries of ^{60}Co for a 10-year period or 1300 kilocuries for a 20-year period. A price of 50 cents/curie is used to estimate the

source cost for encapsulated ^{60}Co initially installed in the MPDI.

If the space for ^{60}Co were doubled in the MPDI source frame, 565 kilocuries would have to be purchased for a 10-year operating period, thus saving 185 kilocuries. Over a 20-year period, a saving of 360 kilocuries would be realized from the additional space.

A hypothetical facility, with a ^{137}Cs source having the same gamma energy yield as the MPDI, would require the purchase of 1280 kilocuries of ^{137}Cs for a 10-year period or 1575 kilocuries for a 20-year period. Encapsulated ^{137}Cs may be available in the future at a price* of about 12 cents/curie.

Use of a point source to generate a cylinder proved to be a convenient technique for establishing the dose pattern in a system where calculations would be tedious or difficult.

Experimental dose measurements demonstrate the possibility of designing a simple, compact irradiator for installation in shellfish or other processing lines. A cost range of 0.8 to 1.6 cents/lb is predicted for a typical set of conditions when irradiation is used as an additional process step. This compares with a predicted cost of 1.5 to 3 cents/lb for irradiation at the MPDI under a similar set of conditions. (FEM)

*Subsequently the AEC has announced consideration of a price *increase* for ^{137}Cs.—*The Editor*

References

1. P. Miller and R. J. Herbert, Marine Products Development Irradiator, *Isotop. Radiat. Technol.*, **1(4)**: 310-317, 322-323 (Summer 1964).
2. Associated Nucleonics, Inc., Preliminary Design Report for Marine Products Development Irradiator Facility, USAEC Report TID-21359 (AN-144), March 1963.
3. C. M. Lederer, J. M. Hollander, and L. Perlman (Comps.), *Table of Isotopes (6th ed.)*, John Wiley & Sons, Inc., New York, 1967.
4. *Physical Aspects of Irradiation*, National Bureau of Standards, Handbook 85, Superintendent of Documents, U. S. Government Printing Office, Washington, Mar. 31, 1964.
5. J. B. Huff and J. M. Lee, Replacement of Underwater Bearings in a High Radiation Field, *Isotop. Radiat. Technol.*, **5(1)**: 25-28 (Fall 1967).

New Food-Irradiation Publications

Three new publications on food irradiation have been announced recently:

Radiation Processing of Food Products, by L. V. Metlitskii, V. N. [I.] Rogachev, and V. G. Krushchev [in Russian; English translation edited by Martha Gerrard, F. E. McKinney, P. S. Baker (Isotopes Information Center), and Eugen Wierbicki (U. S. Army Natick Laboratories), USAEC Report ORNL-IIC-14, July 1968].

A comprehensive survey of food irradiation in the USSR has been published by the Isotopes Information Center (IIC). The original text, *Radiatsionnaya Obrabotka Pishchevykh Produktov*, was translated on an IBM 7090 computer at the Computing Technology Center, Oak Ridge Gaseous Diffusion Plant, using a program developed at Georgetown University. Staff members of the IIC and one member of the U. S. Army's food-irradiation group edited the translation.

The book, published in the USSR in 1967, presents information on sources of ionizing radiation and types of installations used for irradiating food products in the Soviet Union and data on the effectiveness of the process for specific products—both in general terms and in detailed physiological and chemical terms. Most of the investigations were conducted in the A. N. Bakh Biochemistry Institute of the USSR Academy of Sciences and the All-Union Scientific Research Institute of the Food Canning and Vegetable Drying Industries of the Ministry of the Food Industry of the USSR, although some references are made to other USSR and non-USSR institutes and laboratories.

The book is divided into five sections. The first three sections consider the usual categories of radiation treatment based on the kind of specific effect: sprout inhibition, insect disinfestation, and prevention of microbial spoilage. The other two sections cover the wholesomeness and food value of irradiated food products and the sources and facilities available for radiation processing of food.

In addition to sprout inhibition, the authors discuss the effect of radiation on the resistance of potatoes and other vegetables to microorganisms and on the chemical composition of the foods. Under radiation

disinfestation, the authors discuss the disinfestation of specific foods, i.e., grains, food concentrates, dried fruits, dried potatoes, and dried and smoked fish. A major portion of the section on the prevention of microbial spoilage is devoted to the effects on microorganism radiation resistance of dose rate, oxygen, protective substances, sensitizing substances, water, temperature, radiation source, and combinations of these. The effects of radiation on the organoleptic properties of foods and ways to prevent undesirable changes are covered. Individual foods are treated under general categories of foods—meats, fish and fish products, and fruits. The section on wholesomeness and food value of irradiated foods is concerned with the possible occurrence of induced radioactivity and with the toxicity and carcinogenicity of irradiated foods. The food values of vitamins, proteins, carbohydrates, and lipids are treated in detail. After a lengthy description of the current USSR irradiators, as well as of some foreign facilities, the book concludes with an 82-entry bibliography of USSR studies on irradiation of foods.

Preservation of Bread by Means of Gamma Radiation, by G. Stehlik and K. Kaindl (in German; English translation by P. S. Baker, USAEC Report ORNL-IIC-15, July 1968).

Because of the scarcity of literature dealing with radiation processing of bread, the appearance of this publication is a welcome addition. It was originally presented at a seminar on Mar. 21, 1967, and was translated into English to make the information more readily available to workers in the field.

Food Preservation by Irradiation. A second edition of *Food Preservation by Irradiation,* one of the booklets in the USAEC's Understanding the Atom Series, is now available. This second edition is an extensive and complete revision of the earlier version, having been expanded from 36 to 57 pages.

While the revision includes an updating of old material, the expansion is essentially new material. The many different irradiators developed by the USAEC since the booklet last appeared in 1964 are described, and the new information, especially on cost-benefit relationships, that has accumulated since 1964 is incorporated. Some of the changes in the food-irradiation program on fruits are evidenced by the dropping of figs and the adding of pears, papayas, and mangoes in the second edition.

The first two documents are available for $3.00 each from the Clearinghouse for Federal Scientific and Technical Information, National Bureau of Standards, U. S. Department of Commerce, Springfield, Va. 22151. Single copies of the third one are available free from the Division of Technical Information Extension, P. O. Box 62, Oak Ridge, Tenn. 37830. (FEM)

Section

IV

Isotopes and Radiation Technology

Process Radiation Development

Study of Radiation Processing

By D. S. Ballantine*

Abstract: *A report of a study group on the current status of radiation processing of chemicals in the United States is given. Prepared by the U. S. Atomic Energy Commission's (USAEC) Division of Isotopes Development (DID) at the request of the Joint Committee on Atomic Energy, the objectives, survey procedures, and descriptions of commercial radiation processes are presented. General observations are given on the potential of, company attitudes toward, patent policies for, and the role of DID-supported development work in radiation processing. The need for better university, management, and public information programs, dissemination of radiation processing engineering and design data, and AEC–industry cooperative projects is stressed.*

At the request of the Joint Committee on Atomic Energy, the USAEC's Division of Isotopes Development (DID) has made a study of the radiation processing of chemicals in the United States. In this study the term "radiation processing" is restricted to the use of ionizing radiation from either radioisotope or machine sources for effecting useful changes in chemical systems. The object of the study was two-fold: (1) to determine the status of industrial interest and investment in radiation processing and (2) to determine the present attitude of industrial companies regarding radiation processing.

Although there has been a continuing active effort in radiation processing for almost 20 years, it was in 1959 that DID started support of research and development in this area. The total funding for radiation processing operations supported by DID through FY 1968 was approximately $12 million, of which about $4 million was for support of radiation-source development engineering. Thus actual DID support for radiation processing research and development in chemical systems was approximately $8 million.† Brookhaven National Laboratory (BNL) has expended the major portion of DID effort, with the balance of the projects being distributed among universities, research institutes, and industrial companies.

Simultaneously, there have been continuous research and development programs supported by American industrial companies. The study group estimates that during this period the corresponding industrial support was $50 to $75 million. In the early years, radiation processing was heralded as the remedy for many industrial chemical problems; elaborate research facilities were built; and radiation was quickly tried in the hope that it would be the solution to long-standing problems. When quick success failed to materialize, enthusiasm faded and subsequent industrial attitudes toward radiation processing were quite varied. In some companies there has been a continuing significant research and development effort, while in others, radiation has been completely phased out of the research programs. Recent years have been marked by the entry of new companies into process radiation or research and development. The newer entries, profiting from earlier experiences, have approached radiation processing with a much more realistic and sophisticated attitude.

The total current private industrial expenditures for radiation processing research and development is difficult to determine, but it is certainly considerably greater than the current annual $1.4 million expended by DID, which estimates the current annual industrial

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†See appended bibliography of reports of DID-sponsored work on process radiation.

support to be $10 to $15 million. Known processes resulting from this research and development effort and the companies involved are:

1. Cross-linking of polyethylene films to produce special wire coatings, insulating materials, and heat-shrinkable packaging film and bags of improved physical properties: Raychem Corp.; W. R. Grace & Co., Cryovac Division; Electronized Chemicals Corp., a subsidiary of High Voltage Engineering Corp.; and International Telephone and Telegraph Corp.

2. Preparation of specialty copolymers for battery separators that meet Department of Defense requirements: RAI Research Corp. and High Energy Processing Corporation.

3. Synthesis of new graft copolymer fibers with improved crease-resistance and soil-release properties: Deering Milliken & Co., Inc.

4. Production of wood–plastic combinations with improved surface properties and high esthetic appeal: Lockheed-Georgia Company and The American Novawood Corporation. (These suppliers have achieved a limited success in developing a market for their materials.) Atlantic Richfield Co. recently announced plans for large-scale production of wood–plastic flooring.

5. Synthesis of ethyl bromide: Dow Chemical Co.

6. Controlled degradation of Polyox, a polyethylene oxide polymer: Union Carbide Corporation.

7. Curing of surface coatings by machine irradiation of specially formulated, monomer-based lacquers and paints: Developed by Ford Motor Company as the Electrocure process and licensed to Boise Cascade Corp. for treating plywood sheets and siding; radiation curing is also being promoted by Radiation Dynamics, Inc., as the Dynacote process.

The dollar volume of sales of such products has been variously estimated to be about $100 million annually. The annual growth rate in recent years for radiation processing of chemicals and plastics has been 20 to 25%.

Survey Procedures

For making the survey, a list was compiled which the study group felt constituted a representative cross section of industrial chemical companies in terms of size, production interests, and levels of investment in radiation processing research and development work. Also included were research institutes, producers of radiation equipment, and companies that have facilities for radiation research and/or production work on a contract basis.

Some of the companies were visited, and, wherever possible, discussions were held at the level of director, manager, or vice presidents of research. Otherwise, discussions were held with senior research personnel responsible for the radiation research programs. The discussions frequently included four or five of the company personnel representing various research, engineering, and marketing interests.

In other cases, information was obtained by a series of telephone interviews in which specific questions were discussed.

General Observations

Radiation Processing Potential

The potential of radiation processing cannot be measured solely by the number of operating processes. In a significant number of cases, companies have initiated work with radiation and obtained promising laboratory results. But at this stage the research effort was refocused on more classical initiator systems which duplicated the results of radiation research. Further development was then restricted to the classical methods for which extensive experience existed. This is a recurring theme. Seldom does a company, upon discovering a new product by classical techniques, attempt to determine the ability of radiation to duplicate the synthesis.

In other instances, radiation processes have been developed through the pilot-plant stage and then abandoned because of nonradiation factors, e.g., market size, market lifetime expectancy, or market deficiencies—i.e., inability to integrate the product into an existing marketing and sales structure. It is recognized that these considerations are equally applicable to all new processes. The important fact is, however, that radiation has been demonstrated to be a competitive initiator system for chemical reactions.

A specific case involved a much publicized radiation process (Esso) for producing biodegradable detergents. The process, developed through the pilot-plant stage, was efficient, trouble free, and promising as a large-scale industrial process. However, simultaneously, a partially biodegradable detergent, linear alkyl benzene sulfonate (LAS), was developed which, though inferior in biodegradability, largely satisfied existing pollution-control codes. The impact of LAS on the anticipated market was sufficient to halt plans for full-scale production by the Esso radiation process, although Esso has indicated a willingness to discuss licensing with interested parties. If pollution codes are

tightened, interest in this radiation process could be reactivated.

A second case involved the radiation synthesis of a flocculating agent with extremely high efficiency for a specific industrial process application. It is significant that this material could be produced only by a radiation technique. Since the present definable market is limited, the unit cost cannot be made competitive with less efficient but cheaper alternative flocculants. It is felt that, if the market could be expanded threefold, a large-scale radiation process would probably be born.

Similar marketing considerations also thwarted radiation processes in at least three other instances.

Another factor, which critically affects the development of a radiation, or any new process, is the relative size of the market and the company involved. A typical case encountered during this survey was at a large petroleum company. The company found an interesting new radiation-produced product in the laboratory but failed to continue development because the market for the product was too small for a large company to consider. The company's staff felt that a smaller company in the same situation might have proceeded with further development.

In summary, although radiation has demonstrated an ability to compete with classical initiator systems, a number of processes have failed to reach fruition because of (1) instinctive company preference for established alternatives; (2) failure of a market to develop or because the markets were too transient or too small; and (3) the company's market and sales capability was not geared to handle the new product. Of course, these factors apply equally to nonradiation processes.

Company Attitudes Toward Radiation Processing

There is a wide spectrum of attitudes toward radiation among companies with closely related product interests. This is particularly obvious if one surveys a number of petroleum- or rubber-producing companies. In the early 1950's, largely as a result of Esso's lead, most of the major oil companies invested significant amounts of capital funds in radiation facilities. Today, at least one company has stopped all work; some operate at an annual level of about 5 man-years, but only two were reported to be involved in any specific, intensive development.

A similar situation prevails among the rubber companies. At least one reported never having done any "in-house" research, another had given away its radiation source, but a third maintains a continuing annual 4 to 5 man-years of effort.

Some companies, faced with their own lack of success and the general paucity of successful radiation processes, have concluded that radiation processing will always be of marginal value and have abandoned it for other alternatives in the always present competition for research and development dollars. On the other hand, some companies continue to justify research and development on the basis of "spin-off" benefits that have accrued. The general attitude in these companies is that radiation represents another basic mode of initiating chemical reactions and, therefore, merits continued attention.

Lack of Radiation Knowledge

There is a lack of knowledge of radiation fundamentals and the status of radiation processing. After years of continuing work on radiation processing, and in spite of various conferences, symposiums, and technical-society publications, some responsible personnel at levels as high as research director are still unaware of even the fundamental principles of radiation. There is an even broader lack of awareness of present work in radiation processing, costs and availability of radiation sources, and the existence of companies that offer facilities and services in radiation technology and processing.

DID-Supported Work

DID support has stimulated much of the work now under development and previously completed. It is recognized that DID sponsorship of research and development projects in process radiation has been mainly responsible for development of industrial interest in process radiation. In addition to the general stimulation, a number of the specific successful processes are the direct extension of early work supported by DID, e.g., (1) The present Ford Electrocure process is closely related to early work done at BNL on the radiation curing of polyester formulations. Ford's contribution lies in the development of an improved electron gun, a fast-curing coating formulation, and efficient use of energy at high radiation dose rates. (2) The Deering Milliken work on wash–wear fibers is an extension of earlier graft copolymer work supported by DID at various laboratories. (3) The present development of radiation-synthesized wood–plastic combinations is a direct extension of earlier DID

efforts. A number of the companies surveyed had strong interests in or were extending work supported in the recent past by DID. This was true particularly for low-temperature emulsion polymerization, ethylene polymerization and copolymerization, organometallic synthesis, and graft copolymers.

There is a general recognition by industrial companies of an essential DID role in supporting work on the more exploratory aspects of process radiation. However, the extent to which DID should support pilot-plant and demonstration programs provoked a more varying response. The general consensus was that good development work at this stage requires an intimate knowledge of marketing and production economics, as well as engineering experience, which is found only in the realm of competitive industrial companies.

Industrial Needs

A more sophisticated and realistic attitude toward radiation processing is emerging. Companies which are now evidencing interest in radiation for the first time and those taking a new look realize that radiation is not a panacea and that success in its use requires capitalization on the unique characteristics of radiation.

Engineering Data. There is a desire for more data on engineering design and costs. The lack of data on source and equipment design, radiation shielding, and costs tended to drive companies to nonradiation alternatives. Some companies thought that AEC should take greater initiative in getting radiation engineering data into reference handbooks.

Patent Policies. There is a general feeling that patent policy restrictions had inhibited valuable industrial participation in the AEC process radiation program. Some companies, particularly smaller ones with limited research budgets, felt they would like to do research and development work under AEC sponsorship but could not seriously undertake this work without a patent incentive. They indicated that their intimate knowledge and experience in specific areas of technology made them particularly well equipped to make valuable contributions to process development.

There was general agreement that, while BNL and other DID-supported laboratories have done excellent work on research and preliminary engineering studies, they are not well suited for detailed process development because of lack of direct industrial experience.

Specific Observations

In addition to the general observations noted above, there were some very specific items of interest:

1. One company that provides radiation services has been irradiating material, on a 24-hr/day schedule, for an undisclosed company. By the end of 1967, it had irradiated about 750 thousand lb.
2. Another company has been selling, for about a year, a chemical whose synthesis involves a radiation step. The production level is reported to be greater than that of the Dow Chemical Co.'s ethyl bromide process. The company has chosen not to disclose even the fact that a radiation process exists.
3. One company is planning and a second has constructed a pilot plant to produce polyethylene by a radiation process. Each represents an initial investment of about $500 thousand.
4. Several companies offering research and development services are planning to expand their irradiation services [Nuclear Materials and Equipment Corp. (NUMEC), Gamma Process Company, Inc., and The American Novawood Corporation] in response to increasing industrial interest in radiation processing.
5. There is a high level of interest by two companies in radiation-initiated emulsion polymerization for latex paint manufacture.
6. One major rubber company has plans for irradiation of 250 thousand lb of a major product.

Conclusions

The present level of industrial interest and investment in radiation research and development is at least as high as during the peak years of the 1950's. Interests vary considerably—from the use of radiation as a general, broad-base research and development tool to situations where radiation is used to pursue very specific company objectives.

The great disparity of prevailing industrial attitudes toward radiation as a processing tool has a number of causes. Some of the companies were "oversold" initially by exaggerated claims for irradiation. Personnel responsible for fruitless ventures are understandably reluctant to "risk" radiation a second time. Another factor that affects company attitudes toward irradiation involves the training and personality of personnel who make company decisions. Essential ingredients for the development of a vigorous radiation program are well-trained and aggressive proponents at both the research and administrative levels.

Actually, the negative attitudes and lack of success in some companies are not surprising. It is possible to cite instances where personnel designated to become the company's radiation "experts" were not well qualified for the responsibility. This suggests an a priori prejudiced subjective judgment that radiation would probably not be successful.

There is in some circles a radiation prejudice, and, like most prejudices, this is fed by lack of education and experience. Any new technique or process that is to replace existing technology encounters considerable inertia, but process radiation encounters even more. Many company research directors have felt that radiation processes are expensive, hazardous, burdened with labor and public relation problems, costly to insure, and difficult to license. The significant problem here is that these conclusions were reached without detailed knowledge of the facts or even sources of factual information. Such a situation emphasizes the need for broader educational efforts.

Need for Educational Program

There is a great need for a better-planned and integrated educational program which will reach all elements that may influence the growth and acceptance of a process radiation technology. This is probably the single most promising area for future emphasis.

University Training. There are both general and specific opportunities for advancing education in process radiation technology in colleges and universities. Few graduate schools are training chemists who can capably direct industrial radiation programs. Graduate school radiation chemists frequently develop an attitude that research on problems of industrial radiation chemistry is not "pure" and that those who pursue it are not "true radiation chemists." This form of bias, of course, exists also in many other areas of chemistry.

The problem in engineering is more acute because there is an even greater paucity of mechanical and chemical engineers with radiation training. As indicated earlier, this deficiency impedes the smooth progression of a radiation process from the laboratory to pilot plant. In a general sense there is a need to incorporate into chemical and mechanical engineering curriculums the knowledge of radiation initiation and radiation treatment as unit processes. Additionally, there is a need to provide support at the graduate level for research on process radiation problems. At present, there is so little funding for radiation engineering projects in universities that most professors, in the interests of achieving personal recognition, are forced to conduct research in alternative areas where research funds are available. It is essential that these needs be recognized and support be developed for the training of radiation chemists and engineers vitally needed for the development of a process radiation industry.

Management Education. There does not now exist a sufficiently aggressive informational program that meets the specific needs of industrial management charged with making decisions related to process radiation.

Public Education. The general public continues to have erroneous opinions relative to nuclear safety, and this constitutes a factor in management decisions on potential radiation processes.

Radiation Data Dissemination. Many of the basic radiation engineering and design data that have been developed are not so readily available for use as data related to nonradiation technology. This fact, too, tends to influence decisions for adoption of the nonradiation alternatives.

Significance of DID-Supported Work

DID-supported work has had and continues to have a significant effect on promoting industrial process radiation research and development. As indicated earlier, DID work has had, in selected cases, an immediate influence on processes—wood–plastic combinations and ethylene polymerization, for example. In other instances the stimulation has been less direct but is nonetheless evident.

In an industrial company there is always a lag of at least 5 years between a research discovery and a production process. In DID-supported research and development, the lag is necessarily longer because of the need to cross the additional government–industry interface.

There has also been a most significant DID contribution to all radiation processing technology as a result of the continuing BNL work on radiation source technology, radiation shielding, and dosimetry.

Government–Industry Cooperative Effort

The field of radiation processing would be assisted if arrangements could be made for greater AEC–industry cooperative projects. The limitation at this time is the reluctance of industry to participate because of what it considers to be a highly restrictive

patent policy. Industry does not seem to be cognizant of the fact that mutually satisfactory agreements can be and have been negotiated. Industry is also largely unaware of the opportunity for personnel training through guest appointments for work at the BNL High Intensity Radiation Development Laboratory. (REG)

Bibliography

The following bibliography lists all the reports that the editors of *Isotopes and Radiation Technology* have been able to identify that have been published under the Division of Isotopes Development's process radiation program. Progress reports for periods less than a year have been excluded because the work was later reported in completion reports.

L. B. Adler, Heat Transfer Study of Cobalt-60 Shipping Container, Progress Report No. 1, USAEC Report BNL-657, Brookhaven National Laboratory, 1961.

L. B. Adler, Heat Transfer Study of Cobalt-60 Shipping Container, Progress Report No. 2, USAEC Report BNL-684, Brookhaven National Laboratory, 1961.

H. C. Andersen, G. Cohn, C. D. Keith, and P. N. Rylander, AEC Research and Development Report on Effect of Irradiation of Solid Catalysts, USAEC Report NYO-2836, Engelhard Industries, Inc., 1960.

A. A. Armstrong, Jr., and H. A. Rutherford, Applications of Nuclear Radiation and Radioisotopes to Textile Materials and Processes, Annual Report for the Period November 1, 1959–November 1, 1960, USAEC Report NCSC-2477-1, North Carolina State College, School of Textiles, 1960.

A. A. Armstrong, Jr., and H. A. Rutherford, Modification of Textile Fiber Properties by Gamma Radiation, Annual Report for the Period November 1, 1960–November 30, 1961, USAEC Report NCSC-2477-6, North Carolina State College, School of Textiles, 1962.

A. A. Armstrong, Jr., and W. K. Walsh, Modification of Textile Fiber Properties by Radiation-Induced Graft Polymerization, Annual Report for the Period November 1, 1961–October 31, 1962, USAEC Report NCSC-2477-10, North Carolina State College, School of Textiles, 1962.

A. A. Armstrong, Jr., W. K. Walsh, and H. A. Rutherford, Modification of Textile Fiber Properties by Radiation-Induced Graft Polymerization, Final Report for the Period November 1, 1958–October 31, 1963, USAEC Report NCSC-2477-11, North Carolina State College, School of Textiles, 1963.

A. A. Armstrong, Jr., and W. K. Walsh, Radiation-Induced Oxidation of Hydrocarbons, USAEC Report ORO-634, North Carolina State University, Department of Textile Chemistry, 1965.

D. S. Ballantine and L. E. Kopp, Bibliography of Technical Publications and Reports of the Radiation Division, Brookhaven National Laboratory, 1951–1962 Inclusive, USAEC Report BNL-789, Brookhaven National Laboratory, 1963.

D. S. Ballantine and L. E. Kopp, Bibliography of Technical Publications and Reports of the Radiation Division, Brookhaven National Laboratory, January 1963–June 1965 Inclusive, USAEC Report BNL-943 (Suppl. to BNL-789), Brookhaven National Laboratory, 1965.

B. S. Bernstein and G. Odian, A Study of the Mechanism of Radiation-Induced Gelation in Monomer–Polymer Mixtures, Report for Period May 1, 1961–October 31, 1963, USAEC Report RAI-329, Radiation Applications, Inc., 1963.

B. S. Bernstein, J. Lee, and G. Orban, Upgrading of By-Product Polypropylene by Radiation Techniques, Final Summary Report for the Period February 1, 1964–January 31, 1965, USAEC Report RAI-3333-1, Radiation Applications, Inc., 1965.

H. V. Boenig, Investigation of the Pyrolytic Products of Irradiated Cigarettes, Final Report, USAEC Report ORO-633, Spindletop Research, 1965.

W. H. Burrows, W. J. Corbett, P. E. Gaffney, and C. Orr, Jr., Effects of High Energy Ionizing Radiation on Colloidal Systems and Suspensions, Final Report, March 1, 1962–October 31, 1963, USAEC Report TID-20067, Georgia Institute of Technology, Engineering Experiment Station, 1963.

R. W. Carter, R. C. Palmer, and W. V. Willis, Cesium-137 Research Irradiator, Final Report, April 1, 1959–March 31, 1961, USAEC Report SRO-46, Georgia Institute of Technology, Engineering Experiment Station, 1961.

J. E. Christian and W. F. Bousquet, Radioisotopes and Nuclear Techniques in the Pharmaceutical and Allied Industries, USAEC Report TID-11215, Purdue Research Foundation, 1960.

C. H. Collins, H. T. Cullinan, C. C. Thomas, Jr., and S. Weller, Development of a Radioactive Pebble Bed Chemical Reactor System, Final Report: Phase I, USAEC Report NYO-3718-1, Western New York Nuclear Research Center, Inc., 1966.

P. Colombo and M. Steinberg, Cobalt-60 Gamma Irradiation of Uranium and Hydrocarbons, Radiation Processing Report No. 5, USAEC Report BNL-753, Brookhaven National Laboratory, 1962.

P. Colombo, M. Steinberg, and D. Macchia, Cobalt-60 Gamma-Ray Induced Copolymerization of Ethylene in the Presence of Other Monomers, USAEC Report BNL-7134, Brookhaven National Laboratory, 1963.

P. Colombo and M. Steinberg, Copolymerization of Ethylene Induced by Cobalt-60 Gamma Radiation, USAEC Report BNL-6849, Brookhaven National Laboratory, 1963.

Columbia University, Radiation Synthesis of Artificial Kidney Membranes, Annual Report for the Period February 1, 1964–January 31, 1965, USAEC Report NYO-3329-2, 1965.

W. J. Corbett and J. H. Burson III, Delamination of Kaolin by High-Energy, Ionizing Radiation, Final Report, April 1, 1959–March 31, 1961, USAEC Report SRO-57, Georgia Institute of Technology, Engineering Experiment Station, 1961.

Division of Isotopes Development, Potential Requirements for Fission Products as Heat and Radiation Sources. Also includes Appendix: Economic Comparison of the Use of Fission Products and Other Energy Sources in Power Applications, USAEC Report TID-20432, 1964.

Division of Isotopes Development, Process Radiation Development Program Summaries, Second Annual Contractors Meeting, May 28–29, 1962, Held at Brookhaven National Laboratory, 1962, USAEC Report TID-7645, 1962.

Division of Isotopes Development, Process Radiation Development Program Summaries, Third Annual Contractors Meeting, April 22–23, 1963, Held at Brookhaven National Laboratory, 1963, USAEC Report BNL-790, 1963.

Division of Isotopes Development, Process Radiation Development Program Summaries, Fourth Annual Contractors Meeting, June 18–19, 1964, Held at Brookhaven National Laboratory, 1964, USAEC Report BNL-874, 1964.

Division of Isotopes Development, Process Radiation Development Program Summaries, Fifth Annual Contractors Meeting, October 7–8, 1965, Held at Brookhaven National Laboratory, 1965, USAEC Report BNL-949 (also USAEC Report CONF-651064), 1965.

Division of Isotopes Development, Process Radiation Development Program Summaries, Sixth Annual Contractors Meeting, September 26–27, 1966, Held at Germantown, Maryland, 1966, USAEC Report BNL-50042, 1967.

L. W. Elston and W. H. Burrows, Physical Properties and Structural Characteristics of Polymers Resulting from "Post-Effect" Polymerization, Final Report, March 1, 1962–October 31, 1963, USAEC Report TID-20072, Georgia Institute of Technology, Engineering Experiment Station, 1963.

Emerson Radio and Phonograph Corp., A Management Evaluation of Massive Radiation for Industrial Utilization, USAEC Report AECU-4240, 1959.

Emerson Radio and Phonograph Corp., Utilization of Radiation for the Conversion of Grain to Chemicals, Task No. 2, 1959.

J. C. Evans, M. Douglas, D. M. Egan, P. R. McDonald, C. H. Collins, and J. Knopp, Commercialization Studies on Wood–Plastic Combinations Process, Final Report, USAEC Report NYO-3569-1, Western New York Nuclear Research Center, Inc., 1965.

A. M. Feibush, Research on Applications of Krypton-85 and Other Radioactive Isotopes, Final Report, USAEC Report NYO-2906-1, Air Reduction Co., Inc., Central Research Laboratories, 1964.

A. M. Feibush, Research on Applications of Radiation-Induced Reactions in Gases, Final Report, USAEC Report NYO-3334-1, Air Reduction Co., Inc., Central Research Laboratories, 1965.

P. Y. Feng, Radiation Synthesis Involving Halogen Compounds, Final Report, May 1, 1960–October 31, 1965, USAEC Report COO-578-3-32, Illinois Institute of Technology, IIT Research Institute, 1966.

A. F. Fentiman, Jr., R. E. Wyant, J. L. McFarling, E. J. Kahler, and J. F. Kircher, Radiation-Induced Formation of Organometallic Compounds with Particular Emphasis on Organotins, USAEC Report BMI-1753, Battelle Memorial Institute, 1966.

A. F. Fentiman, R. E. Wyant, R. D. Steinmeyer, D. A. Jeffrey, J. F. Kircher, and E. J. Kahler, Radiation-Induced Formation of Organotin Compounds, USAEC Report BMI-1713, Battelle Memorial Institute, 1965.

J. H. Frankfort and K. M. Black, Engineering and Evaluation Study for the Manufacture of Wood–Plastic Composites, USAEC Report KLX-1876, Vitro Engineering Company, 1966.

J. H. Frankfort, S. Haram, Jr., and S. Wallach, An Experimental Gamma Irradiator for Hospital Sterilization, USAEC Report NYO-9432 (AN-119), Associated Nucleonics, Inc., 1960.

J. H. Frankfort, S. Haram, Jr., and S. Wallach, An Industrial Gamma Irradiator for Medical Supplies, USAEC Report NYO-9433 (AN-120), Associated Nucleonics, Inc., 1960.

S. A. Goldblith, Summary of Findings on Strontium-90 and Cesium-137 Process Control Devices, Final Report, USAEC Report NYO-9181, Massachusetts Institute of Technology, 1960.

M. A. Golub and R. L. Walrath, Radiation-Induced Reactions in Swollen Polymers, Annual Report, USAEC Report TID-18126, Stanford Research Institute, 1962.

W. W. Graessley and J. H. Zufall, Fission Product Applications Using Gaseous Beta Sources, Task II, USAEC Report NYO-9485, Air Reduction Co., Inc., 1961.

R. E. Greene and P. S. Baker (Eds.), Proceedings of Information Meeting on Irradiated Wood–Plastic Materials Held at Conrad Hilton Hotel, Chicago, Illinois, September 15, 1965, USAEC Report ORNL-IIC-7, Oak Ridge National Laboratory, 1966.

E. I. Heiba, The Effect of Electrophilic Additives on the Radiolytic Products of Organic Chain Reactions, Annual Report, USAEC Report NYO-10194, Little (Arthur D.) Inc., 1961.

K. M. Held and R. J. Goldman, Synthesis of Semiconductor Materials by Radiation Induced Reactions, Final Report, USAEC Report NYO-9864 (also USAEC Report TRG-132), TRG, Inc., 1962.

K. Held and R. J. Goldman, Synthesis of Semiconductor Materials Using Nuclear Radiation, Final Report, May 1, 1959–October 31, 1962, USAEC Report NYO-10472, TRG, Inc., 1963.

S. K. Hellman, Use of Radiation Techniques for the Inhibition of Bacterial Growth in Liquid Media, USAEC Report KLX-1872, Vitro Engineering Company, 1965,

B. Y. Howard and W. S. Koski, The Radiation Chemical Reactions of Olefins with Methanethiol and with Methylsilanes and of Dimethylamine, USAEC Report JLI-3655-200, Johnston (William H.) Laboratories, Inc., 1968.

F. D. Iannazzi, P. L. Levins, F. G. Perry, Jr., and R. S. Lindstrom, Technical and Economic Considerations for an Irradiated Wood–Plastic Material, Final Report, USAEC Report TID-21434, Little (Arthur D.) Inc., 1964.

Johnston (William H.) Laboratories, Inc., Report on Industrial Radiation and Its Prospects, 1959.

Johnston (William H.) Laboratories, Inc., Basic Studies in Radiation Technology, Final Report, November 2, 1959–October 31, 1961, USAEC Report JLI-650-8-8, 1962.

Johnston (William H.) Laboratories, Inc., Basic Studies in Radiation Technology, Final Report, November 1, 1961–April 30, 1966, USAEC Report JLI-2901-75, 1966.

Johnston (William H.) Laboratories, Inc., Training Experiments in Radiation Chemistry, Vol. I, Experiments; Vol. II, Theory, USAEC Report JLI-650-2-1, 1960.

W. H. Johnston, Special Report on Ozone Formation in the Radiolysis of Oxygen, USAEC Report JLI-2901-0-1, Johnston (William H.) Laboratories, Inc., 1963.

J. A. Kent, A. Winston, and W. Boyle, Modification of Wood with Polymers Formed by High Energy Radiation, USAEC Report TID-18907, West Virginia University, 1963.

J. A. Kent, A. Winston, and W. R. Boyle, Preparation of Wood–Plastic Combinations Using Gamma Radiation to Induce Polymerization; Effects of Gamma Radiation on Wood, Annual Report, November 1, 1961–October 31,

1962, USAEC Report ORO-600, West Virginia University, Engineering Experiment Station, 1963.

J. A. Kent, A. Winston, and W. R. Boyle, Preparation of Wood–Plastic Combinations Using Gamma Radiation to Induce Polymerization. Interim Report, USAEC Report ORO-612, West Virginia University, Engineering Experiment Station, 1963.

J. A. Kent, A. Winston, W. R. Boyle, W. Loos, and J. E. Ayres, Preparation of Wood–Plastic Combinations Using Gamma Radiation to Induce Polymerization, USAEC Report ORO-628, West Virginia University, Engineering Experiment Station, 1965.

J. A. Kent, W. E. Loos, W. R. Walters, W. R. Boyle, G. W. Taylor, and A. Winston, Preparation of Wood–Plastic Combinations Using Gamma Radiation to Induce Polymerization, Topical Report on Impregnation of Wood with Acrylic Monomers, USAEC Report ORO-2945-4, West Virginia University, Engineering Experiment Station, 1966.

J. A. Kent, A. Winston, W. R. Boyle, and G. B. Taylor, Preparation of Wood–Plastic Combinations Using Gamma Radiation to Induce Polymerization, Interim Report, USAEC Report ORO-2945-7, West Virginia University, 1967.

J. A. Kent, G. B. Taylor, W. R. Boyle, and A. Winston, Radiation Induced Polymerization of Vinyl Monomers Impregnated in Wood, USAEC Report ORO-2945-8 (also CONF-661201-3), West Virginia University, 1966.

J. Kivel, Organometallic Radiation Dosimetry Systems, USAEC Report NYO-2979-34, Atlantic Research Nuclear Corp., 1965.

J. Kivel, M. S. Chatterton, and R.J.F. Palchak, Microbial Inhibition Systems Employing Radiation (MISER), USAEC Report NYO-3736-2, Atlantic Research Corp., 1966.

C. N. Klahr, Fabrication of Semiconductor Devices by Neutron Transmutation Doping, Annual Report, December 1961–December 1962, USAEC Report NYO-10088, Fundamental Methods Associates, 1962.

C. N. Klahr and M. S. Cohen, Fabrication of Semiconductor Devices by Neutron Transmutation Doping, Annual Report, December 1962–December 1963, USAEC Report NYO-3124-1, Fundamental Methods Associates, 1963.

C. N. Klahr and M. S. Cohen, Fabrication of Semiconductor Devices by Neutron Transmutation Doping, Annual Report, June 1966, USAEC Report NYO-3124-2, Fundamental Methods Associates, 1966.

J. A. Knight, Jr., and C. J. Bryan, Radiation-Induced Cyanogenation of Organic Compounds, Annual Technical Status Report No. 1, May 1, 1960–March 31, 1961, USAEC Report SRO-51, Georgia Institute of Technology, State Engineering Experiment Station, 1961.

J. A. Knight, Jr., Radiation-Induced Cyanogenation of Organic Compounds, Final Report, May 1, 1960–February 28, 1963, USAEC Report SRO-78, Georgia Institute of Technology, State Engineering Experiment Station, 1963.

O. A. Kuhl, L. Galanter, S. Zwickler, and B. Manowitz, Theoretical and Experimental Studies of Sodium Irradiator Systems, USAEC Report BNL-567, Brookhaven National Laboratory, 1959.

G. Lellouche and M. Steinberg, Industrial Chemical Production by Chemo-Nuclear Processes with Special Reference to the Production of Fixed Nitrogen, USAEC Report BNL-574, Brookhaven National Laboratory, 1959.

P. L. Levins, E. I. Heiba, and D. D. McRitchie, The Effect of Electrophilic Additives on the Radiolytic Products of Organic Chain Reactions, Final Report, USAEC Report NYO-10025, Little (Arthur D.) Inc., 1963

Little (Arthur D.) Inc., Radiation: A Tool for Industry, USAEC Report ALI-52, 1959.

Little (Arthur D.) Inc., Outlook for Industrial-Process Uses of Megacurie Gamma Irradiators, Supplement to Report on *Radiation: A Tool for Industry*, USAEC Report ALI-52 (Suppl.), 1959.

W. E. Loos and J. A. Kent, Fastening of Wood–Plastic Combinations, Topical Report, USAEC Report ORO-2945-9, West Virginia University, Engineering Experiment Station, 1967.

M. Luttinger, F. A. Sliemers, Jr., J. F. Kircher, and R. I. Leininger, Development of High-Dielectric-Constant, Low-Dielectric-Loss Polymers Utilizing Gamma Irradiation, USAEC Report BMI-1678, Battelle Memorial Institute, 1964.

B. Manowitz, R. H. Bretton, L. Galanter, and F. X. Rizzo, Computational Methods of Gamma Irradiator Design, USAEC Report BNL-889, Brookhaven National Laboratory, 1964.

B. Manowitz and D. M. Richman, The Use of Sodium Reactor Coolant as a Source of Gamma Radiation, USAEC Report BNL-3703, Brookhaven National Laboratory, 1958.

R. Markle, F. A. Sliemers, J. F. Kircher, W. B. Gager, and R. I. Leininger, Radiation-Induced Changes in Methacrylate Polymers and Their Application in Postirradiation Graft Polymerization, USAEC Report BMI-1695, Battelle Memorial Institute, 1964.

E. A. Mason, R. C. Reid, and N. J. Stevens, The Effect of Electron Irradiation Prior to Reaction on the Activity of a Semiconductor Catalyst, USAEC Report NYO-9217, Massachusetts Institute of Technology, Department of Chemical Engineering, 1961.

J. L. McFarling and J. F. Kircher, Experiments on Enhanced Catalytic Activity from Internal Irradiation of Catalysts, USAEC Report BMI-1620, Battelle Memorial Institute, 1963.

D. J. Metz and D. S. Ballantine, Gamma Radiation Induced Ionic Vinyl Polymerization, USAEC Report BNL-11344 (also CONF-670516-1), Brookhaven National Laboratory, 1966.

D. J. Metz, Introductory Lectures on the Properties, Detection, and Chemical Effects of High Energy Radiations, USAEC Report BNL-748, Brookhaven National Laboratory, 1962.

H. S. Miller, J. H. Zufall, and W. G. Marancik, Fission-Product Applications Using Gaseous Beta Sources, Task I, USAEC Report NYO-2607, Air Reduction Co., Inc., 1960.

A. Muller, F. Rizzo, and L. Galanter, Solar Cell Dosimeter, Report No. 1, USAEC Report BNL-721, Brookhaven National Laboratory, 1962.

North Carolina State College, Textile Research Center, Applications of Nuclear Radiation and Radioisotopes to Textile Materials and Processes, Annual Report for the Period November 1, 1958–November 1, 1959, USAEC Report ORO-305, 1959.

G. Odian, M. Sobel, R. Klein, and T. Acker, The Accelerating Effect of Additives on Radiation-Induced Graft Polymerization, Annual Report for the Period December 16, 1959–December 15, 1960, USAEC Report NYO-2530, Radiation Applications, Inc., 1961.

G. Odian, T. Acker, E. Ratchik, M. Sobel, and R. Klein, The Accelerating Effect of Additives on Radiation-Induced Graft

Polymerization, Final Report, USAEC Report RAI-301, Radiation Applications, Inc., 1962.

G. Odian, W. F. Oliver, L. J. Friedman, T. Acker, E. Peirre, J. Foltin, and E. Ratchik, A Comparison of Radiation-Induced Graft Copolymerization Utilizing Electron Accelerators and Isotope Sources as Radiation Initiators, Final Report, July 18, 1960–October 17, 1961, USAEC Report NYO-9420, Radiation Applications, Inc., 1961.

G. Odian, B. S. Bernstein, J. Schaefer, L. J. Friedman, and J. Kelly, A Study of the Mechanism of Radiation-Induced Gelation in Monomer–Polymer Mixtures, Final Report, July 13, 1961, USAEC Report NYO-2481, Radiation Applications, Inc., 1961.

G. Odian and B. S. Bernstein, A Study of the Mechanism of Radiation-Induced Gelation in Monomer–Polymer Mixtures, Summary Report for May 1, 1961–April 30, 1962, USAEC Report RAI-306, Radiation Applications, Inc., 1962.

G. Odian and B. S. Bernstein, A Study of the Mechanism of Radiation-Induced Gelation in Monomer–Polymer Mixtures, Report for May 1, 1961–October 31, 1962, USAEC Report RAI-311, Radiation Applications, Inc., 1962.

M. J. Oestmann, E. J. Kahler, G. A. Lutz, and J. F. Kircher, Radiation-Induced Nitration of Hydrocarbons, USAEC Report BMI-1495, Battelle Memorial Institute, 1961.

A. Oltmann and O. A. Kuhl, Fabrication of BNL Standard Cobalt-60 Source, USAEC Report BNL-848, Brookhaven National Laboratory, 1964.

M. Prince, Combined Effects of Ultra-High Pressure and Radiation on Organic Monomers and Polymers, Final Report for March 1, 1963–October 31, 1964, USAEC Report NYO-3185-1, New York University, 1964.

Proceedings of the International Symposium on Radiation-Induced Polymerization and Graft Copolymerization, November 29–30, 1962, Battelle Memorial Institute, USAEC Report TID-7643, 1962.

R. A. V. Raff, The Grafting of Organic Polymers onto Inorganic Systems, Final Report, USAEC Report RLO-2043-2, Washington State University, 1968.

L. Rebenfeld and H. J. White, Jr., Study of the Industrial Potential of Radioisotopic Methods in the Textile Industry, Period Covered: April 1959–June 1960, USAEC Report NYO-2640, Textile Research Institute, 1960.

Z. Reyes, Radiation-Induced Reactions in Swollen Polymers, Final Report, USAEC Report SRIA-120, Stanford Research Institute, 1963.

P. J. Roberts, W. L. Bridges, and A. O. Burford, Industrial Evaluation of Radiation Processed Wood–Plastic Composites, USAEC Report ORO-3415-1, Lockheed Nuclear Products, 1966.

C. A. Rohrmann, Irradiated Wood–Plastic Materials Commercialization in the Pacific Northwest and Great Lakes Regions, USAEC Report BNWL-261, Pacific Northwest Laboratory, 1966.

H. M. Rosenstock, C. R. Mueller, M. B. Wallenstein, M. L. Vestal, A. Tory, D. Rivers, and W. H. Johnston, Ion-Molecule Reactions, USAEC Report JLI-650-3-7, Johnston (William H.) Laboratories, Inc., 1959.

H. Sadowski, K. Held, and J. Beckerley, Nuclear Radiation Processing of Optical Maser Materials, USAEC Report NYO-3330-11, Radioptics, Inc., 1966.

H. Sadowski, K. Held, and J. Beckerley, The Investigation of the Use of Nuclear Radiation in the Preparation of Optical Maser Materials, Final Report, USAEC Report NYO-3330-18, Radioptics, Inc., 1968.

J. Silverman, J. J. Weinstock, H. W. Chandler, and E. J. Henley, The Technology and Applications of Large Fission Product Beta Sources, USAEC Report NYO-2503, Radiation Applications, Inc., 1961.

F. A. Sliemers, Jr., J. F. Kircher, R. Lieberman, R. Markle, W. B. Gager, M. Luttinger, and R. I. Leininger, Radiation-Induced Changes in Polymers and Their Influence on Graft Polymerization, USAEC Report BMI-1610, Battelle Memorial Institute, 1963.

F. A. Sliemers, Jr., J. F. Kircher, W. B. Gager, I. S. Ungar, E. Gülbaran, and R. I. Leininger, Radiation-Induced Graft-Polymerization Studies, USAEC Report BMI-1543, Battelle Memorial Institute, 1961.

Southern Interstate Nuclear Board, Commercialization of the Process of Manufacturing Radiation-Produced Plastic-Impregnated Wood in the Southern Region, USAEC Report TID-22774, 1966.

J. A. Stanfield, The Use of Radioisotopes in the Study of Modifications of Platinum Catalysts, USAEC Report SRO-44, Georgia Institute of Technology, Engineering Experiment Station, 1961.

V. Stannett, J. A. Gervasi, J. J. Kearney, and K. Araki, Radiation-Induced Emulsion Polymerization, Final Report, February 1, 1964–March 31, 1967, USAEC Report TID-24281, Research Triangle Institute, 1967.

V. T. Stannett, D. T. Turner, D. Campbell, Y. P. Castille, H. G. Clark, J. A. Cleveland, J. E. Herz, J. J. Kearney, and J. B. Lando, The Radiation-Induced Polymerization and Copolymerization of Unusual Monomers, Final Report, USAEC Report ORO-637, Research Triangle Institute, 1968.

M. Steinberg and G. Farber, Chemonuclear Fission Fragment Source and Fuel Element Development Progress Report, USAEC Report BNL-827, Brookhaven National Laboratory, 1963.

M. Steinberg and B. Manowitz, The Chemonuclear and Radiation Chemical Process Development at Brookhaven National Laboratory, USAEC Report BNL-8056, Brookhaven National Laboratory, 1964.

M. Steinberg and B. Manowitz, Purification of Mine Drainage Water by a Radiolytically Induced Chain Process, USAEC Report BNL-11220, Brookhaven National Laboratory, 1967.

M. Steinberg, Radiation Processing Report No. 3, Gamma Irradiation of Gaseous CO_2, USAEC Report BNL-665, Brookhaven National Laboratory, 1961.

M. Steinberg, P. Colombo, R. N. Chapman, and G. Adler, Polymerization of Ethylene by Cobalt-60 Gamma Radiation, Radiation Processing Report No. 4, USAEC Report BNL-740, Brookhaven National Laboratory, 1962.

D. S. Thompson and V. Benaroya, Preliminary Design Study for a Sodium Graphite Reactor Irradiation Facility, USAEC Report NAA-SR-3260, Atomics International, Division of North American Aviation, Inc., 1959.

I. S. Ungar, J. F. Kircher, W. B. Gager, F. A. Sliemers, Jr., and R. I. Leininger, A Study of Radiation-Induced Changes in Polymers Leading to Graft Copolymerization as Influenced by Structural Factors, USAEC Report BMI-1511, Battelle Memorial Institute, 1961.

Unidynamics/Phoenix, a Division of UMC Industries, Inc., The Effects of Ionizing Radiation on Colloids, Final Report

Covering May 1964–May 1966, USAEC Report COO-1653-15, in press.

Universal Match Corp., Unidynamics Division, Investigation of the Effects of Ionizing Radiation on Synthetic Detergents, Sewage Colloidal Characteristics and Sewage Sedimentation, Final Report, USAEC Report COO-275, 1963.

Vitro Engineering Company, Preliminary Design Report, Title I on Mobile Gamma Irradiator, USAEC Report KLX-1866, 1964.

L. Voyvodic, Design Studies on Cesium-137 as a Source for High Level Gamma Irradiators, Final Report, June 1, 1959–July 31, 1960, USAEC Report ARF-1150-12, Illinois Institute of Technology, Armour Research Foundation, 1960.

J. I. Wagner and J. S. Milau, Failure of Cobalt-60 Radiation Sources, USAEC Report BNL-50099, Brookhaven National Laboratory, 1968.

M. B. Wallenstein, H. M. Rosenstock, and W. H. Johnston, Fundamental Considerations of Radiation Technology, USAEC Report JLI-650-3-5, Johnston (William H.) Laboratories, 1959.

Washington State University, Nuclear Applications to the Wood, Paper, and Pulp Industry, USAEC Report TID-7704 (also CONF-624), 1964.

J. J. Weinstock, A. J. Karalis, and R. Dell'Abate, The Design of Beta Irradiators—Research and Commercial Scale, USAEC Report RAI-203, Radiation Applications, Inc., 1962.

J. J. Weinstock, Y. Miron, and P. M. Krishna, The Development of Large Beta Radiation Sources and Their Applications, USAEC Report RAI-202, Radiation Applications, Inc., 1963.

R. E. Wyant, J. L. McFarling, J. F. Kircher, and E. J. Kahler, Radiation-Induced Formation of Organotin Compounds, USAEC Report BMI-1654, Battelle Memorial Institute, 1963.

J. Y. Yang, H. T. Cullinan, and C. C. Thomas, Jr., Development of a Radioactive Pebble Bed Chemical Reactor System, Final Report Phase II, USAEC Report NYO-3864-1, Western New York Nuclear Research Center, Inc., 1968.

P. M. Yavorsky, N. J. Mazzocco, and E. Gorin, Beta Radiation-Induced Telomerization of Methanol with Ethylene, Final Report for the Period November 1, 1964–April 30, 1965, USAEC Report NYO-2978-44, Consolidation Coal Company, 1965.

P. M. Yavorsky and E. Gorin, Evaluation of Beta Radiation as a Hydrogenation Catalyst, Interim Technical Status Report, March 1, 1962–October 31, 1963, USAEC Report NYO-2978-34, Consolidation Coal Company, 1964.

P. M. Yavorsky and E. Gorin, Beta Radiation Processing at Rigorous Conditions, USAEC Report CONF-187-77, Consolidation Coal Company, 1963.

P. M. Yavorsky, N. J. Mazzocco, and E. Gorin, Evaluation of Beta Irradiation of Pressurized Reactions, Interim Technical Status Report, November 1, 1963–October 31, 1964, USAEC Report NYO-2978-42, Consolidation Coal Company, 1965.

G. A. Young (Comp.), Utilization of Gross Fission Products, A Bibliography of Unclassified Report Literature, USAEC Report TID-3046, Division of Isotopes Development, 1954.

G. A. Young and R. E. Allen (Comps.), Utilization of Fission Products, A Bibliography of Selected Unclassified Literature, USAEC Report TID-3046 (Suppl. 1), Division of Isotopes Development, 1955.

G. A. Young (Comp.), Radiation Application and Fission Product Utilization, A Bibliography of Selected Unclassified Literature, USAEC Report TID-3046 (Suppl. 2), Division of Isotopes Development, 1959.

(RHL)

Extracts from a Report* on

Grafting of Organic Polymers onto Inorganic Substances

Summarized by R. E. Greene

Abstract: *Extracts from a report (RLO-2043-2) by R. A. V. Raff, summarizing and discussing critically the work reported in the literature on the grafting of organic polymers onto inorganic substances, are presented. The text of the complete report is supported by 245 references, documenting the development of this new branch of polymer science from 1946 through 1967. The report should be useful in generating and planning new research in polymer grafting where atomic radiation can perform in a manner distinctly different from, and superior to, that of more conventional methods. Extracts include introduction to the subject, discussion of the mechanism of adhesion of polymers to inorganic substrates, radiation polymerization onto inorganic substances, and summary and conclusions.*

In work sponsored by the USAEC's Division of Isotopes Development, R. A. V. Raff, at Washington State University in Pullman, has prepared a literature review that organizes, summarizes, and discusses critically the work on grafting organic polymers onto

*R. A. V. Raff, The Grafting of Organic Polymers onto Inorganic Substances, USAEC Report RLO-2043-2, Washington State University, Pullman, Jan. 30, 1968.

inorganic substances, published during the period 1946 to December 1967. Certain events that may be considered preliminary to mixed grafting, as well as those processes where mixed grafting might be suspected without having been proved or claimed, are also discussed. By its very nature and approach, mixed grafting reaches into the domain of materials science by establishing relations between polymeric, metallic, and ceramic materials. Thus the review deals with a field of current interest, which is still in development and which promises to become a new branch of polymer science.

The attitude of many polymer scientists toward the grafting of organic polymers onto inorganic materials has been either outspoken skepticism or enthusiastic acceptance, and these divergent attitudes are reflected in many of the publications. While the writer admits leaning toward the enthusiastic group—else he would not have undertaken the work—he nevertheless has attempted an unbiased presentation, with carefully drawn conclusions.

If one were to keep strictly within the scope of the review title, only those instances would be discussed where covalent, or chemical, bonding forces are involved or suspected. In the literature search it soon became evident, however, that consideration must also be given to the functioning of any other bonding forces; their actions, in many cases, must be considered as being preliminary to the phenomenon of true grafting, while in others no further involvement can be assumed.

A general absence of information permits the imagination of the enthusiastic investigator to suspect reactions involving polymer grafting onto inorganic materials of playing an important role in processes where such reactions have never been claimed, still less proved. At least one chapter in the review is devoted entirely to this extension of the subject, while occasional references are found in most of them.

It is easier to establish the absence of any grafting than to prove grafting where it might be suspected. Such proof, at times quite difficult even when dealing with combinations of organic polymers, becomes more difficult when the system involves mixed organic–inorganic structures. Consequently no well-planned outline can be proposed for the characterization of organic–inorganic graft copolymers. The required overall approach is more like that of detective work, different for different systems and involving a combination of several individual approaches. In view of this, various techniques applied by researchers in the field are summarized in one chapter.

The more careful one is in arriving at conclusions, the fewer conclusions he can make. The chapter on techniques of graft characterization is proof in point since it is among the shortest. Nevertheless, the author indicates that there are numerous instances of established grafting of polymers onto inorganics, depending on the materials involved and on the particular conditions under which these grafting reactions are conducted.

Despite these positive statements, many systems reported in the literature require further study and substantiation of some of the claims made. Also, many of the processes mentioned in this review, where mixed grafting might be suspected, are deserving of investigation for theoretical as well as practical reasons.

All in all, this is still an open field for both fundamental and applied research that reaches into disciplines as remotely connected as prebiology. The primary objective of the review will have been fulfilled if it helps to generate and plan new research, particularly in the use of high-energy radiation as a means of inducing mixed graft copolymerization.

Included in the original report, but not in these extracts, are chapters on glass-fiber-reinforced laminates; grafting on glass fibers treated with $TiCl_4$; adsorption of polymers on inorganic substances; pretreatment of clay surfaces; catalyzed polymerization of gaseous monomers at liquid–gas interfaces; polymer formation on electrical contacts and sliding metals; polymer grafting onto carbon black; polymer grafting onto inorganic substances in polymer processing; biological significance of polymer grafting onto inorganics; and techniques of graft characterization.

Polymerization and Grafting Mechanisms

Graft copolymers are polymers with branched chains—a main chain (backbone chain) with chemically different side chains. These copolymers thus represent molecularly homogeneous mixtures of two polymers, one forced upon the backbone chain of the other by the primary bonding of the side chains; otherwise, spontaneous separation of the components would occur, since different polymers are only rarely compatible and thus mutually soluble.

Graft copolymers, with the backbone as well as side chains composed of organic molecules, have been widely studied[1-3] for their scientific interest. They are also of technical importance since they combine the properties of two, often incompatible, polymers. Several methods have been established for the preparation of organic graft copolymers:

1. An active site capable of initiating graft copolymerization may be formed on a polymer via a chain transfer step, both in free-radical and ionic systems.

2. Grafted chains may be formed by the radical attack on an unsaturated backbone chain.

3. Polymers containing chemically reactive or functional groups along or at the ends of a backbone chain may lead to graft copolymerization by reacting with monomers or polymers.

4. Photochemical free-radical reactions may be used to prepare graft copolymers.

5. Ionizing radiation may lead to graft copolymers, either by mutual irradiation of a monomer–polymer mixture or by prior formation of polymeric peroxides. The latter, formed when a polymer is irradiated in air, may then be decomposed thermally in the presence of the monomer.

6. Mechanical action, such as mastication or milling (shearing action), vibromilling, exposure to ultrasonic waves, spark discharge, or application of freezing and thawing cycles, may break primary bonds in a polymer chain. Subsequent or simultaneous reactions of the resulting radicals with oxygen, vinyl monomers, or polymers may lead to graft copolymerization.

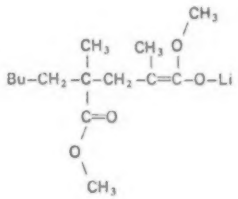
A wide variety of graft copolymers has been prepared by these methods, involving not only synthetic but also natural polymers—cellulose, starch, proteins, natural rubber, etc.—serving as the backbone chain. It is quite difficult at times to ascertain that true grafting has really taken place, particularly if the graft copolymer still contains homopolymer. Methods of characterization of graft copolymers, which have much in common with those used in the fractionation of a homopolymer, have been outlined.[3,4]

In recent years a considerable amount of the literature has suggested that organic polymers can also be attached to inorganic compounds by essentially the same methods of grafting described, i.e., by primary or secondary bonding. However, more specifically, the terms "grafting" or "mixed grafting"* will be applied only in instances where primary bonding is involved. The terms will not be used to describe attachment by secondary bonding forces or by dispersion effects (random, fluctuating polarization). (For a lucid presentation of bonding forces in materials, see the recent book edited by Pask.[5])

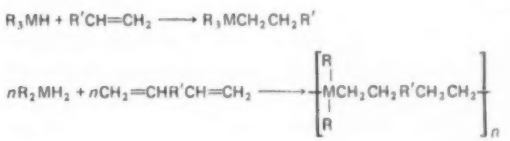
*The term "mixed graft" is used in the review as an alternative term to describe grafts of organic polymers on inorganic substances. One might consider introducing it officially into polymer science, with its proper definition.

While in some publications no proof or only insufficient evidence is given that validates the claimed character of the product—true polymer grafts onto inorganics—it is entirely feasible that structures of this type may be obtained under appropriate reaction conditions. A few examples of this type of grafting are given.

As one example, metal–polymer bonds are claimed by Korotkov, Mitsengendler, and Krasulina[6] to be formed in the polymerization of methyl methacrylate with butyllithium, leading to complexes of the structure



The addition reactions of organometallic hydrides to carbon–carbon multiple bonds have been applied to the preparation of a number of different types of organometallic, step-growth polymers:



This type of reaction has been used for the synthesis of organotin and organosilane polymers, with Sn or Si indicated by M in the above reactions. These and recent achievements in the preparation of semi-inorganic (organic–inorganic) polymers by step-growth polymerization reactions have been summarized by Lenz.[7]

Ziegler–Natta catalysts of various types, covalently bound to polymer chains, have been prepared[8] by reacting pendent organometallic groups on polymer molecules with $TiCl_3$ or $TiCl_4$. The appropriate organometallic groups have been introduced into the polymer chain either by copolymerizing a monomer containing an organometallic group, such as *p*-tributyltinstyrene,[9] or by adding an organometallic hydride to a double bond in a preformed polymer.[10] These catalysts were then used for the graft polymerizations of ethylene, propylene, and styrene.

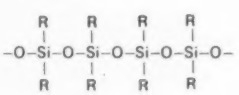
Metal-bonded polymer chains are formed during polymerization with catalysts of the Ziegler and Ziegler–Natta types. Natta, Giuffré, and Pasquon[11]

found that at the end of the polymerization reaction most of the polymer chains are bound to aluminum.

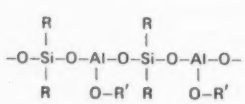
Friedlander and Oita,[12] in introducing the concept of polymerization by ion radicals in a chemisorbed monomer layer (with movement of the growing center through the chemisorbed monomer layer replicating the catalyst surface), assumed that a metal–carbon bond is formed at each end of the olefin double bond. Similar assumptions are made for the Ziegler–Natta catalysis in mechanisms proposed by Natta,[13] Patat,[14] and Uelzmann.[15] These assumptions are in all instances supported by experimental data.

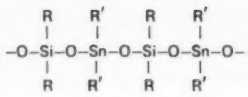
The introduction of inorganic elements, such as Si, P, Mg, Al, Ti, and Sn, in either the backbone chain or the substituents of an organic polymer, has been frequently proposed. As one example, Adrova, Koton, and Klages[16] describe the preparation of organometallic polymers with tin atoms in the backbone chain. These polymers, formed by reacting SnH_2 or $(C_6H_5)_2SnH_2$ with ethylenedimethacrylate or *p*-phenylene dimethacrylate, were prepared primarily with the intent of obtaining products with increased thermal stability.[17] The backbone of these polymers is made from some combination other than carbon–carbon bonds, but to make the polymer elastic, resinous, or plastic, the organic side chains are maintained. Block copolymers of polysiloxanes and poly(alkylene oxides), in which the polymer blocks are bound to each other by Si–C bonds, have been patented by Dow Corning Corp.[18]

Well-known, representative organometallic polymers[19] are the silicone polymers or polysiloxanes of the general formula

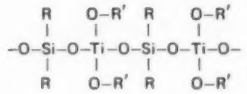


where R can be CH_3, C_2H_5, C_6H_5, or CF_3. An inherent tendency of the chains of these thermally quite stable polymers to form into rings, especially at higher temperatures, leads to a progressive softening of the materials and eventually to a complete loss of toughness, strength, and elasticity. This tendency can be reduced by the introduction of other elements, e.g., Al, Ti, or Sn, into the backbone chain to yield polymers of the general form

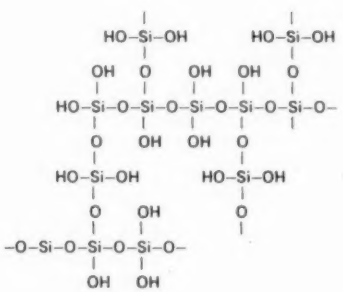




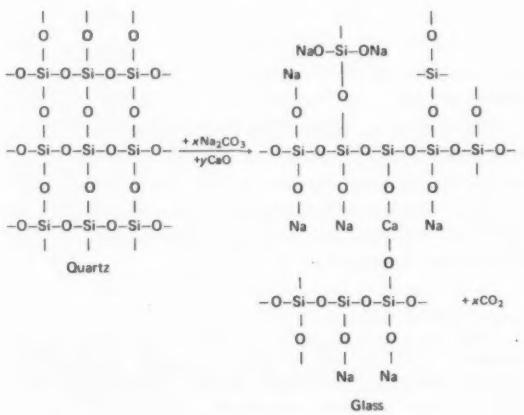
and



Replacement of part of the silicon atoms in the silicone chain by metal atoms is also found in inorganic glasses. Thus, in structure and properties, the silicones range between the organic and the inorganic polymers (glass, asbestos, silicic acid, etc.). Polymeric silicic acid, obtained, e.g., by hydrolysis of $SiCl_4$ and removal of the formed HCl by dialysis,[20] is representative of an inorganic polymer. Further condensation leads to branched and cross-linked polysilicic acids that deposit as gels:

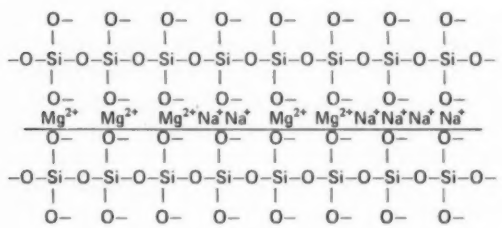


The formation of inorganic glasses may be considered as "Umkondensation" (transcondensation) of quartz sand by the action of soda and lime:



Connection has thus been established with mineral substances, where both reactions in and on inorganic solids and the reaction mechanisms based on structure have been intensely studied.[21] Furthermore, inorganic chemistry, as pointed out by Van Wazer,[22] is to a great extent the chemistry of high polymers. However, while the structural chemistry of inorganic compounds is generally equilibrium controlled so that the relative proportions and kind of reaction products are more readily interpreted in terms of equilibrium constants, the reactions of organic chemistry are essentially kinetically controlled. Also, low-lying orbitals occur in bonding of inorganic polymers, and addition polymerization in inorganic chemistry, exclusive of carbon, seldom if ever involves opening of a multiple bond as is the case in vinyl polymerization, for example.

Many inorganic polymers have been studied in greater detail, and the schematic diagram below[23] shows the structure of silicate rock minerals, e.g., pyroxene, amphibole, or asbestos:



The atoms of the individual chains of these inorganic polymers, like the atoms of organic polymers, are joined by covalent bonds. However, while the adjacent chains of organic polymers are held together by the weaker van der Waals forces, dipoles, or hydrogen bonding, the adjacent chains of such inorganic polymers are held together by ionic bonds. Many differences in physical and mechanical behavior may then be explained by these differences in the dominant bonding forces.

If one considers the atomic structures of such materials as layer-lattice silicates, sodium chloride, graphite, or a metal (body-centered cubic), the possibilities of attaching organic polymer chains to such inorganic structures may be visualized, and unusual combinations of properties might be expected from the resulting materials. In this connection, Collman and Pitt[24] are currently attempting to synthesize macromolecules with backbones comprising entirely intermetallic bonds. This work is expected to lead to a technology of intermetallic polymers because these macromolecules could be structurally modified in many ways, and their physical and structural properties could be adapted to meet specific requirements.

Since the objective of this review is to consider interactions of organic polymeric and inorganic materials, one may begin by investigating what occurs when their surfaces come into close contact. If one assumes that interaction occurs at all, adhesion (to be defined) will precede all other events, one of which may be mixed grafting.

Adhesion of Polymers to Inorganic Substrates

Adhesion can be defined as the attraction between two contiguous surfaces; it may lead to either *adsorption* or *absorption.*

The complexity of the phenomenon of adhesion is reflected in the vast number of publications dealing with its various aspects as well as in attempts[25] to distinguish between different types of polymer adhesion on the basis of the physical states of the adhesive and the adherent. Recently the polymer concept, in adhesives in particular, has been extremely well presented and interpreted by Weiss.[26] The "ideal" adhesive of the future, based on the accumulated results of studies in polymer science, has been described by Mark[27] and by Atlas and Mark.[28]

A classification of the phenomenon of adhesion on the basis of the forces responsible was attempted by Voyutskii;[29] he discussed critically the various existing theories. A chemical theory of adhesion applies most closely to instances which, under the terminology of this review, one might consider as grafting. There are physical theories of adhesion, such as the adsorption theory (van der Waals interaction), the diffusion theory (diffusion of long-chain molecules or their segments), the hydrogen-bond-formation theory, and the electrical theory (contact electrification of two dielectrics or of a metal and a dielectric). None of these, however, Voyutskii concludes, can pretend to be universally applicable, nor can adhesion of high polymers to metals be reduced simply to chemical or physical interactions. Different types of adhesion may have very different causes, and, in many instances, the question of the adhesion mechanism is still quite unresolved.

Although only very few points of attachment may be assumed to exist between a clean solid surface and an adhesive, many observations[26] indicate that polymeric materials of varying chemical composition, applied under similar conditions, may yield bonds differing greatly in strength or in resistance to thermal deterioration. In particular, Lewis and Forrestal[30] concluded that acids or electron-accepting groups

generally increase adhesion to metals. Consequently these authors proposed a chemical reaction or transfer of electrons across the interface between the metal surface and the polymer to explain the adhesion mechanism.

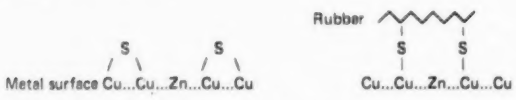
The geometric arrangement of the adhesive molecules on metal surfaces also has a strong effect on the adhesive force between the polymer and the metal. This arrangement and the matching of polymers [polyethylene, poly(vinyl chloride)] and metal surfaces, aided by the use of molecular models, have been described by Taylor and Rutzler.[31] Inadequate matching of the chemical groups in an adhesive with sites on the surface of the substrate is considered to cause inferior adhesion.[32]

According to Huntsberger,[33] wetting and bond formation are synonymous in many cases. The molecular configuration of the polymer in the adhesive is believed to be one of the factors in establishing whether adhesive failure occurs at the interface or through cohesive failure in the adhesive itself.

Two types of interaction can cause high heats of adsorption:[34] the attraction of polar molecules to ionic solids and the process of chemisorption, which may include covalent, ionic, or metallic bonds to the surface. In metal–polymer adhesion, it may be safely assumed that the metal surface is, in fact, an oxide surface.[35]

Covalent interactions between alkyd resins containing organic acids and metals were claimed by Grinsfelder[36] and between vulcanized rubber and brass by DeBruyne,[37] Kaercher and Blum,[38] Buchan and Rae,[39] and Robins.[40] The chemical and technological investigations of Buchan and Rae, combined with the electron diffraction study of Robins, were discussed by Wake.[41]

Robins removed rubber from bonded units by freezing them in liquid nitrogen. Although he found the surface of the brass completely free of rubber, it no longer showed the diffraction pattern of alpha brass; rather, it was covered with a green iridescent film that gave the diffraction pattern of hexagonal CuS, which must be assumed to have formed during vulcanization. Buchan[42] concluded that bonds develop through the formation of Cu_2S, which then reacts with the rubber molecule at points of unsaturation; one can assume a bridge structure consisting of a sulfur atom between Cu_2S and rubber:



Wake[41] admits that the conclusions from his survey of adhesive mechanisms are only tentative. He considers the brass bond to rubber almost certainly to be chemical, but other bonds are probably largely physical. The distinction between chemical and physical adsorption for the isocyanate bond to metals or oxides may not be quite so clear, and both mechanisms may take part.

The adhesion of a large number of elastomers of the most varied nature to the surfaces of various metals was investigated by Voyutskii and Markin.[43] Their data were interpreted as showing that the adhesion of nonpolar elastomers to aluminum and zinc is governed mainly by dispersive forces; that of polar elastomers, by the considerably stronger molecular interactions (dipoles, hydrogen bond) or even by the formation of chemical bonds.

In a subsequent publication, Voyutskii et al.[44] reported an attempt to obtain further information on the nature of the forces determining the adhesion of polymers to metals by investigating the temperature dependence of adhesion and determining the corresponding values of the apparent activation energy. It was concluded from the values for the apparent activation energy of adhesion (characteristic of processes where the action of molecular forces is predominant) that the formation of covalent chemical bonds is not probable for the adhesive–substrate combinations investigated.

Characteristics of peeling as a test for bonding were determined on the basis that polymers, contrary to metals, fluoresce markedly in ultraviolet light. If there remained on the metal a very thin layer of polymer, although invisible to the naked eye or even under a microscope, the surface would show intense fluorescence; with purely adhesive failure of the bonded structure, no fluorescence would be observed.

The assumption that a paint film applied to a metal should differ on opposite sides if the paint film had any special affinity for the metal (or the metal surface oxide) was tested by Tamai and Tanaka.[45] This was done by measuring the contact angle[46] of water on both the air- and the metal-side surfaces of plastic film coatings removed from metal. The plastic used was a methacrylate copolymer that was baked on Cd, Al, Ni, Fe, and Au. Contact angles for the air side were about 74° in all cases, but values on the metal side differed considerably, ranging from 76° for Au to 50° for Cd. The film–metal combinations that showed the larger contact angle difference (increasing in the following order: Au, Fe, Al, Ni, Cd) generally also showed higher breaking strength.

Finally, Spitsyn et al.[47] investigated the effect of high-energy radiation on the adhesion of polyethylene to aluminum. Irradiation of a polyethylene coating on aluminum as well as prior irradiation of polyethylene powder, up to 10 to 15 Mrads, essentially doubled the adhesive force; with still higher irradiation, adhesion deteriorates. The type of polyethylene, as well as the surrounding atmosphere, has considerable effect on the nature of the adhesion. Irradiation of the coating with the base is more effective than irradiation of the powder. Upon examining the various physicochemical processes that occur at the interface as a result of irradiation, one concludes that the improvement in adhesion results from the radiation-chemical oxidation of polyethylene in the contact zone. This oxidation promotes a favorable orientation of the carbonyl groups in relation to the Al_2O_3 film and also improves the flexibility of the macromolecules in the radiation field, thereby increasing the number of contacts between adhesive and substrate.[48]

Spitsyn et al.[47] also considered the possibility that radiation might induce chemical interaction between the polymer coating and the aluminum surface as a result of the reactions

$$RH \xrightarrow{\gamma} R^{\cdot} + H^{\cdot} \quad (1)$$

$$>AlOH \xrightarrow{\gamma} Al{-}O^{\cdot} + H^{\cdot} \quad (2)$$

$$>AlO^{\cdot} + R^{\cdot} \rightarrow Al{-}O{-}R \quad (3)$$

$$>AlOH + R^{\cdot} \rightarrow Al{-}O^{\cdot} + RH \quad (4)$$

$$>AlOH + ROO^{\cdot} \rightarrow Al{-}O^{\cdot} + ROOH \quad (5)$$

$$>AlOH \xrightarrow{\gamma} >Al^{\cdot} + OH^{\cdot} \quad (6)$$

$$>Al^{\cdot} + R^{\cdot} \rightarrow >Al^{\cdot}{-}R \quad (7)$$

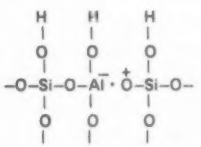
where $R^{\cdot}$ is the polyethylene radical and $Al{-}O^{\cdot}$ the aluminoxy radical. Reactions 1, 2, and 4 lead to a growth in the number of $R^{\cdot}$ and $Al{-}O^{\cdot}$ radicals. Since radicals recombine more slowly in crystalline than in amorphous polyethylene, there is an accumulation of $R^{\cdot}$ and $ROO^{\cdot}$ radicals. The yield of $Al{-}O^{\cdot}$ radicals by reaction 2 is not very high, but their concentration may rise appreciably from reaction 4. The recombination of $R^{\cdot} + Al{-}O^{\cdot}$ radicals then gives a strong Al–O–R bond.

It has thus been shown that chemistry, morphology, and geometry of organic polymeric and inorganic surfaces will determine whether, and what kind of, bonding forces may be generated on close contact. Only when these surfaces carry atoms or groupings of atoms with a strong tendency to mutually react can the formation of primary bonds be expected. This sequence of events is particularly well demonstrated in structures where glass fibers are incorporated into resins to achieve reinforcement and where surface treatments may supply the mutually reactive groupings required for mixed grafting.

Radiation Polymerization on Inorganic Substances

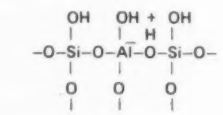
Ionizing radiation, as shown by Kazanskii, Pariiskii, and Voevodskii,[49,50] affects the surface properties of silica gel. When thermally pretreated silica gel samples were irradiated in vacuum at the temperature of liquid nitrogen with gamma rays from a ^{60}Co source, and then investigated by electron spin resonance (ESR), scission of bonds in surface hydroxyl groups of the silica was found to take place, leading to formation of hydrogen atoms stabilized in crystal-lattice vacancies in the surface layer. The adsorbed hydrogen atoms thus formed were highly reactive and interacted chemically with oxygen and ethylene at −150 and −120°C, respectively. The hydrogen-atom signal from these irradiated samples was accompanied by a complex signal caused by silica gel defects. However, based on an evaluation of the signal intensities, not all defects formed during irradiation were assumed to be detectable by ESR.

Further work on the nature of surface defects in irradiated silica gel was reported by Pariiskii, Mishchenko, and Kazanskii,[51] who studied the mechanism of the irreversible adsorption of hydrogen. The structure of the surface color centers, formed by irradiation and bonded to an aluminum impurity in samples of irradiated silica gel, was established from the optical and the ESR spectrums of the samples. The structure is represented schematically as



Thus these surface color centers are positively charged holes stabilized on the oxygen atom next to the impurity aluminum atom; they are also the centers of

the irreversible adsorption of hydrogen. When adsorption takes place, the hole is destroyed and the initial surface structure of the nonirradiated silica gel is restored; this is shown schematically as



Griffiths, Owen, and Ward[52] had established much earlier that the color of irradiated quartz samples is proportional to the intensity of the ESR signal.

The effects of radiation on metals, the association of their changes in physical properties with atomic displacements, and the production of interstitial atoms and vacancies have been summarized and discussed by Seitz.[53] Similar considerations apply to graphite.

In further studies, Kazanskii et al.[54-56] evaluated the ESR spectrums of organic radicals adsorbed on the surface of silica gel. For example, ethane was adsorbed at room temperature on silica gel in amounts less than a monolayer and then cooled to −196°C and irradiated at this temperature with ^{60}Co gamma rays. The ESR spectrum of the ethyl free radicals formed showed that the adsorbed radical is connected with the surface at only one end. The study of the spectrum obtained while the specimen was heated to above liquid-nitrogen temperature showed that the adsorbed radicals were extremely stable. According to these and other properties derived from the spectrum, one concludes that the adsorbed radicals should be similar to free radicals in gaseous and liquid phases, and that their free electrons should not participate to any considerable extent in the formation of a chemical bond with the surface. The strength of their bond with the surface is about 3 to 5 kcal/mole, they are very reactive, and they are assumed to play a role in heterogeneous catalysis.

The rate of the ionic polymerization of isobutylene, induced at low temperatures by high-energy radiation, was found by Worrall and Pinner[57,58] to be increased considerably when the reaction takes place in the presence of certain inorganic compounds, notably oxides of high surface area. These included ZnO, CaO, MgO, SiO_2, and $NaHCO_3$. While the increase in polymerization rate is assumed to be caused by an increase in the rate of initiation, no reference is made to any type of bonding between the polymer formed and the inorganic surface.

Vlasov et al.,[59] in an attempt to have a semiconducting polymer supported by a strong base, studied the possibility of gas-phase polymerization by irradiating acetylenic monomers (phenylacetylene) and acrylonitrile on glass fiber. Glass fiber was chosen since its thermal stability permitted the subsequent heat-treatment at 500°C necessary to obtain a semiconductor. The glass fibers were irradiated in a special apparatus in the absence of air, which permitted the fibers and the monomer reservoir to be maintained at different temperatures. A considerable amount of polymer (up to 20% of the fiber weight) was deposited on the fiber. This process is described as grafting, although no proof is given. A similar study was also reported by Polak et al.[60]

The apparatus used in these experiments (Fig. 1) has been described by Vlasov[61] and consists of two connecting ampules, joined at one end to a vacuum pump. Liquid or liquefied monomer is placed in one of the ampules, which is surrounded by a cooling bath. The glass fiber is inserted into the other ampule, and this ampule is then placed in a thermostatically controlled enclosure fitted with an aluminum-foil window for irradiation. Polymerization experiments are conducted, after evacuation of the system, by heating each ampule to a predetermined temperature.

The same apparatus was also used in the polymerization studies of Tsetlin et al.,[62] who claimed that poly(methyl methacrylate), polyacrylonitrile, and poly(vinyl chloride) were grafted to carbon black, powdered silica gel, and MgO at 40 to 50°C with beta irradiation. Methyl methacrylate polymerized intensively on BeO, but not on ZnO. It is believed that the reactions proceed by a radical mechanism and that the polymer chain is initiated by the oxygen ion radical formed at the crystal-lattice surface.

In still another paper, Tsetlin[63] showed that, when mineral powders (MgO, SiO_2) were treated with gaseous monomers by X rays or by electrons from a linear accelerator in the apparatus shown in Fig. 1, polymers insoluble in specific solvents resulted, indicating the absence of homopolymer. A soluble polymer residue obtained upon dissolving the inorganic base in hydrogen fluoride affords proof of true grafting.

It is pointed out that radiation grafting is thus basically different from mechanical grafting; by effective dispersion of mineral substances in monomers, a large amount of homopolymer is formed.[64] The specific surface (area) of mineral oxide was increased 15 to 20% in the described radiation grafting. For MgO the yield of graft copolymers was proportional to the square root of the dose intensity, which is considered to indicate radical initiation.

Polystyrene grafted onto silica gel and then sulfonated gave materials that combine the advantages of

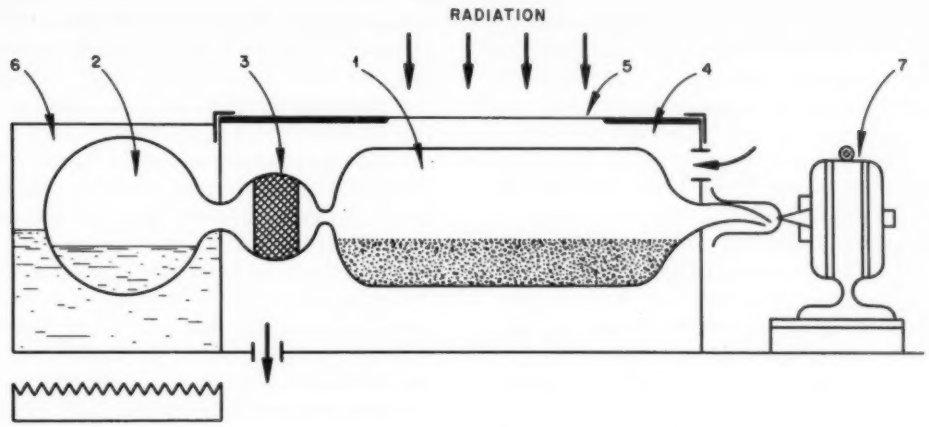


Fig. 1 Graft polymer synthesis by the irradiation of "beds" of solids in the presence of monomer vapors: (1) reaction ampule; (2) monomer ampule; (3) glass filter; (4) controlled-temperature enclosure; (5) aluminum-foil window; (6) monomer bath; (7) vacuum pump.

ion-exchange resins and mineral sorbents. These materials show high sorption rates, especially from dilute solutions; they are capable of both ionic and molecular sorption as well as of sorption of colloidal particles; and they swell very little in water. Similar sulfonated products, based on grafted polystyrene (21%) and poly(acrylic acid) (25%), were also prepared and described by Egorov et al.[65]

The kinetics of X-ray-initiated grafting of acrylonitrile onto glass fibers was investigated by Morozov et al.[66] On porous glass fibers the initial polymerization rate is higher than that on nonporous fibers. After a certain period, when the pores become filled with polymer, the rates are nearly the same. Curves of initial polymerization rate vs. monomer vapor pressure show that the rate reaches a limiting value at a vapor pressure at which the amount of adsorbed monomer also reaches a limiting value. The rate of this polymerization is thus determined by the concentration of the monomer in the adsorbed layer. With porous fibers, the polymerization rate is linearly proportional to the dose rate; with nonporous fibers, to its square root. The graft polymerization is shown to proceed by a free-radical mechanism and is probably induced by the oxygen radical and terminated on porous fibers by interaction of macroradicals with the walls of the pores.

The kinetics of polymerization from the gas phase of acrylonitrile onto a mineral powder, initiated with gamma radiation from ^{60}Co, was investigated by Berezkin, Kolbanovskii, and Kyasimov;[67] they also referred to the work of Tsetlin.[63] The purpose of the experiments was to modify the properties of a brick material widely used as the support in gas–liquid chromatography. The apparatus used was similar to that described by Vlasov.[61] The quantity of graft polymer, reported in weight percent of adsorbent, was determined as a function of irradiation time for constant dose rate and constant vapor pressure of acrylonitrile:

Time, hr	Graft polymer formed, wt.% of adsorbent
0	0
1	4.7
2	8.6
3	11.0
9	23.9

The kinetics of this grafting process is claimed to be in good agreement with that of adsorption on a heterogeneous surface.

Uskov et al.,[68] acknowledging that reinforcement of polymers by active fillers depends on sufficiently strong interaction between the solid surface and the bonding polymer, consider it more promising to graft from liquid monomer than from monomer vapor when using radiation polymerization in the presence of a filler.

In the reported study, fillers (silica gel, kaolin, asbestos, glass fiber), which as such do not initiate

polymerization, were sealed with styrene or methyl methacrylate in ampules filled with nitrogen and irradiated with gamma rays from a ^{60}Co source. The amount of homopolymer formed was determined from the refractive index, and that of the grafted (insoluble) polymer from the loss of weight on calcination of the dried filler after all monomers had been extracted with an appropriate solvent.

Silica gel was most active in increasing both the rate of the radiation-induced polymerization and the molecular weight of homopolymer—more so than either kaolin or asbestos. Glass fiber showed no effect. These differences are attributed to differences in the specific surface areas and in the crystalline structures of these fillers.

In surface grafts, polystyrene is believed to be held to the fillers by chemical bonds, while poly(methyl methacrylate) is believed to be adsorbed to a considerable extent by hydrogen bonding as well as by being chemically grafted. This conclusion is derived from the interpretation of observed differences in solubility in different solvents. The yields of homopolymer and of grafted polymer increased with temperature, indicating a free-radical polymerization mechanism. Polystyrene was also grafted onto a layer of poly(methyl methacrylate) previously grafted onto a mineral filler.

Several points are emphasized in Uskov's paper. The increased rate of radiation polymerization in the presence of inorganic additives, also observed by others,[57] probably occurs because radiolysis of the monomer is energetically more favored in the adsorbed state than in bulk.[69] The active radicals produced by hydrolysis are more stable in the proximity of a solid surface;[49] this stability also promotes an increase in the polymerization rate in the presence of mineral additives. The stability of the adsorbed radicals is evidently considerably greater for fillers with a low surface area and is believed to explain the increase in molecular weight observed by Uskov.

Chemical grafting is thus associated with the formation of short-lived defects on a solid surface, followed by the generation of free radicals on this surface.[70] The surface radicals initiate polymerization, and, if the bond with the carbon atom is sufficiently strong, a graft polymer is formed; if the bond is weak, a macroradical or macromolecule detaches itself from the surface to yield a homopolymer. If the polymer is capable of bonding to the solid surface through hydrogen bonding, its higher concentration near the solid surface promotes strong adsorption.[71]

The radicals on the solid surface are, at the same time, chain-termination centers. Therefore, when their concentration is high, the molecular weight and the amount of grafted polymer formed per unit surface area are lower for porous fillers with a high specific surface, such as silica gel, and higher for smooth-surface fillers, such as glass fibers.

Separate experiments showed that the total surface area of the powder sample remains essentially constant in every case up to 11 wt.% of grafted material (relative to weight of substrate). For a low specific surface area of the initial solid-phase sample and high percentages of weight increase, the grafting of acrylonitrile no longer takes place on the actual surface of the inorganic material alone but partially onto already grafted polyacrylonitrile.

The graft copolymerization of vinylpyrrolidone on sheets of poly(dimethyl siloxane) by the mutual irradiation technique, utilizing high-energy electrons, has been described by Yasuda and Refojo,[72] who were working toward a medical application of the grafted rubber. Water was found to accelerate the polymerization and to reduce the penetration of the monomer solution into the polymer. The dependence of the molecular weight of poly(vinyl pyrrolidone) on the radiation dose was determined, as well as the dependence of the penetration of the grafted layer in the poly(dimethyl siloxane). The graft copolymers are hydrophilic in proportion to the amount of polymer grafted onto the silicone.

The location of the graft copolymer was examined by a staining technique with dyes used for staining proteins, e.g., eosin. The depth of penetration was determined after sectioning and was converted to percent of penetration in the total thickness of the grafted sheets. Grafting was found to vary from surface grafting to apparently homogeneous grafting, depending on the water content in the monomer solution and on the radiation dose.

The mechanism of the radiation polymerization of vinyl monomers (styrene, methyl methacrylate) in the presence of mineral additives (ZnO, NiO, MgO, silica gel) was also studied by Kornienko et al.,[73] who concluded that a free-radical mechanism was involved. The increase in polymer yield observed in some instances was related to the catalytic action of the additive, which led to an increase in the rate of chain-formation initiation. Chain formation was initiated in the monomer layer adsorbed on the inorganic additive but continued into the interior of the monomer phase. No reference is made to any polymer grafting taking place onto the oxides.

The practical importance of radiation polymerization onto inorganic substances is reflected in the

patent literature. Organic–organosilicon graft copolymers were prepared by Warrick.[74] Finely dispersed inorganic solids (including silica, silicates, metal halides, oxides of Mg, Ca, Sr, and Ba) were suspended in a mixture of an organopolysiloxane and a vinyl monomer and polymerized with either ionizing radiation (Van de Graaff generator) or a free-radical-initiating organic peroxide or azo compound. Increased thermal stability, stabilization towards hydrolysis, and changes in compatibility and solubility are claimed to be achieved by this process.

An inorganic refractory compound, preferably a finely divided oxide, was exposed to ionizing radiation by Ingersoll[75] and then contacted with the monomer to be grafted. Silica, alumina, zirconia, and aluminum silicates were the preferred substrates, and methyl methacrylate and tetrafluoroethylene the preferred monomers. At least some of the polymer is claimed to be chemically bonded to the inorganic compound, as shown by reduced solubility. The process provides a method of initiating polymerization without directly irradiating the monomer. The inorganic refractory material, normally incapable of initiating polymerization, can be used to initiate polymerization immediately after irradiation; or it can be retained for several days, preferably in the absence of oxygen and at lower temperatures, before being contacted with monomer. Additional fracturing or grinding of the irradiated product will further increase its capability of initiating polymerization.

A new general class of materials, polyorganosilicate graft polymers, with higher resistance to heat and solvents than currently available polymers, is claimed by Nahin.[76] In his experiments, the surface of the clay was "primed" with polyethylene-like tails and bridges by reacting hydrogen clays with hexamethylenediamine or poly(vinyl alcohol). The tails and bridges were then cross-linked to polyethylene by gamma irradiation. This reaction yields a three-dimensional polymer in which the organic chains are claimed to be directly bonded to large silicate sheets, resulting in a product of great strength, excellent thermal stability, and resistance to organic solvents.

One of Nahin's patents[77] describes the preparation of clay fillers that can be incorporated in substantial amounts into thermoplastic polymers, e.g., polyethylene, polypropylene, polystyrene, poly(vinyl chloride), without destroying their tensile strengths and other desirable properties. For the filler, an adduct of a clay with an organic base [oleylamine, poly(allyl amine), 4-vinylpyridine] is prepared; the adduct is incorporated into the polymer; and, after being heat fused, the blend is intensely irradiated with gamma or beta rays to effect cross-linking. In another patent,[78] the organo-clay adduct is made by contacting a hydrogen clay with poly(vinyl alcohol) or poly(ethylene oxide).

Summary and Conclusions

When two surfaces, one of an inorganic substance and the other of an organic polymer, come into close contact, their chemical structures and their geometries decide whether or not bonding forces will be aroused. Bonding, however, may stop short of grafting unless the surfaces carry atoms or groupings of atoms that have a strong tendency to mutually interact; only then will organic (covalent) bonds be formed.

Such a role is apparently being played by the oxide surface layer of metals or by the sulfur atoms in vulcanized rubber. Treatments introducing groups of this kind, such as surface oxidation of metals or the oxidative formation of carbonyl groups of polymer surfaces (both known to improve adhesion), may thus establish or further enforce chemical bonding. High-energy radiation, known to increase bonding between a polymer and a metal, may do this by partial oxidation of the polymer surface. However, there exists the alternative possibility that chemical interaction, induced by the radiation, leads to the formation of metal–oxygen–polymer bonds between the surface of the polymer and the activated metal oxide layer.

Bonding may also be induced by a pretreatment of the surfaces with compounds carrying mutually reactive groups. This is apparently the function of the silane-type coupling agents used in glass-fiber-reinforced laminates. Observed insolubilization of the polymer and the results of infrared absorption studies substantiate the claims of the occurrence of covalent bonding. Moreover, on the basis of the solubility data, more pronounced grafting through silanes was found when the surface-treated glass fiber was added during polymerization than when added to the polymerized resin. Photomicrographs show the grafts as isolated buds on the surface of the glass.

While surfaces may induce the polymerization of monomers, the resultant polymers need not be grafted. Surface treatments can, however, cause these polymerizations to result in mixed grafts.

One instance, particularly well studied, is the pretreatment of glass fibers with $TiCl_4$. In the absence of $TiCl_4$, polymerization of vinyl monomers leads to soluble polymers; with $TiCl_4$ adsorbed on the glass, an insoluble, grafted polymer is formed. The ESR spectrums of the systems consisting of $TiCl_4$, glass, and

monomer suggest the presence of paramagnetic complexes on the surface of $TiCl_4$-treated glass, which function as centers for the grafting of the polymerizing monomer. Again, photomicrographs show the grafts as nodules at isolated places on the glass fiber.

Adsorption may be viewed as adhesion onto a substrate of layers of molecularly dimensioned thickness and may be discussed in an analogous manner. When applied to the problem of polymer–filler interaction, all stages from mere physical inclusion to covalent grafting can be observed, again depending on chemical structure, geometry, and surface morphology, with surface irregularities and dislocations playing an important role. This interaction leads to a reduction of the supermolecular mobility in the adsorbed layer, similar in effect to that of cross-linking. As in adhesion, this need not, and in many instances probably does not, entail polymer grafting, but the same or similar events may be assumed to occur as preliminaries to mixed grafting.

Since surface interactions are governed by both the chemistry and the morphology of the surface, changes in either may be applied to achieve mixed grafting.

There are several ways of changing the surfaces of fillers, particularly of clays, by chemical means. In one type of reaction, use is made of the acidic character of the clay surface; this acidic surface combines with nonpolymeric bases such as amines or their derived compounds. In another, the clay surface is clad with a preformed polymer. While no claims of mixed grafting are made in either instance, treatments that render clay surfaces active for subsequent polymerization reactions may lead to polymer grafts. This treatment has been demonstrated. Vinyl monomers were attached to the clay surface, and these vinyl clays were polymerized by conventional means. In this polymerization, radical initiators were attached to the clay surface and the resultant products (e.g., azo clays) were applied both to induce vinyl polymerization and to propagate vinyl polymers from their surfaces.

For changes, not necessarily identical, in the surface morphology of inorganics, two approaches have been studied in particular. One consists in the establishment of freshly formed surfaces by mechanical attrition, including ultrasonic treatment, of inorganic materials; the other involves the application of ionizing radiation.

Changes in the structure of freshly formed surfaces are indicated by their ESR spectrums. Inorganic surfaces thus form specific centers of electron-donor or free-radical types and become capable of initiating polymerization reactions. These may lead to the formation of mixed grafts if the attrition of the inorganic substance is carried out in the presence of a monomer. In the case of metals, the mechanism proposed for the initiation of the polymerization of vinyl monomers is associated with direct transfer of an electron to the monomer molecule by exceeding the escape energy of the electron. For ionic salts, ionization of defects of the "color center" (*F*-center) type, accompanied by electron emission, is held responsible. Rupture of covalent bonds in mechanical attrition in SiO_2, TiO_2, graphite, and other inorganics, as well as in oxide films on the surface of metals, causes formation of radical centers capable of initiating radical polymerization which may entail mixed grafting.

Organic polymers may be attached on carbon black, with or without previous surface modification of the latter. A graft copolymer of high-density polyethylene and carbon black was prepared by simultaneous mechanical dispersion of the components and was identified as a graft copolymer by electron-microscope and X-ray analysis. The formation of polymer observed on electric contacts and on sliding metals may be interpreted as polymerization on freshly formed surfaces; whether this is accompanied by mixed grafting has not yet been investigated.

The other approach to changing the morphology of inorganic surfaces involves the application of ionizing radiation. This method is occasionally combined with a surface pretreatment with a nonpolymeric or a polymeric compound or with an additional fracturing or grinding step.

Numerous studies have been made of the effects of ionizing radiation on metallic and mineral surfaces and the resulting surface defects. Information on the bonding of organic radicals was obtained in ESR studies of irradiated surfaces on which low-molecular-weight compounds had been adsorbed.

The reported increase in the rate of radiation polymerization in the presence of inorganic additives may be attributed to the fact that the energetics of radiolysis of the monomer favor the adsorbed rather than the bulk state. The resultant surface radicals initiate polymerization. For example, if the bond with the carbon atom is sufficiently strong, grafted polymer is formed; if the bond is weak, a macroradical or macromolecule detaches itself from the surface to yield homopolymer. These possibilities are essentially substantiated by the reported experimental results.

In a number of commercial processes involving energy input into systems consisting of a polymer and inorganic matter, any observed extraordinary performance may possibly be caused by mixed grafting. In

addition to its being of theoretical interest, a study of mixed grafting possibilities may be technologically rewarding.

Polymer grafting onto inorganics may also have played a determining role in prebiological events. A question might be raised as to whether or not certain chemical elements, e.g., magnesium, iron, and trace elements, which are essential parts of living organisms, have been introduced at an early stage of development through organic compounds or polymers that had remained grafted to active centers of inorganic surfaces on which they had formed and grown.

Finally, it is emphasized that the characterization of mixed grafts cannot follow a prearranged scheme but must consist of the assembling of numerous pieces of information. How these have been obtained by researchers in this field is summarized in the report.

References

References marked with an asterisk are not available in the Isotopes Information Center.

1. W. J. Burlant and A. S. Hoffman, *Block and Graft Polymers,* Reinhold Publishing Corporation, New York, 1960.
2. R. J. Ceresa, *Block and Graft Copolymers,* Butterworths, London, 1962.
3. *H. A. J. Battaerd and G. W. Tregear, *Graft Copolymers,* Interscience Publishers, New York, 1967.
4. R. J. Ceresa, The Characterization of Block and Graft Copolymers, in *Techniques of Polymer Characterization,* pp. 231-251, P. W. Allen (Ed.), Butterworths, London, 1959.
5. J. A. Pask (Ed.), *An Atomistic Approach to the Nature and Properties of Materials,* John Wiley & Sons, Inc., New York, 1967.
6. A. A. Korotkov, S. P. Mitsengendler, and V. N. Krasulina, Kinetics and Mechanism of Polymerization of Methyl Methacrylate by Butyllithium, *J. Polym. Sci.,* **53:** 217-224 (1961).
7. R. W. Lenz, *Organic Chemistry of Synthetic High Polymers,* pp. 172-175, 199, Interscience Publishers, New York, 1967.
8. *Ibid.,* p. 719.
9. G. Greber and G. Egle, Anionic Graft Copolymerization. VII. Graft Copolymerization of Ethylene with Tin and Lead Containing Macromolecules and Compounds of the Transition Metals (in German), *Makromol. Chem.,* **64:** 207-209 (1963).
10. R. J. Ceresa, The Physico-Chemical Synthesis of Block Copolymers, *Polymer,* **1:** 397-399 (1960).
11. G. Natta, L. Giuffré, and I. Pasquon, Determination of Al in the Polypropylene Molecule (in Italian), *Atti Accad. Naz. Lincei, Rend., Cl. Sci. Fis., Mat. Natur., Ser. 8,* **25:** 417-423 (1958).
12. H. N. Friedlander and K. Oita, Organometallics in Ethylene Polymerization, *Ind. Eng. Chem.,* **49:** 1885-1890 (1957).
13. G. Natta, I. Pasquon, and E. Giachetti, Kinetics of the Chain Growth and Decomposition Processes During the Stereo-Specific Polymerization of Propylene (in German), *Makromol. Chem.,* **24:** 258-290 (1957).
14. F. Patat and H. Sinn, The Course of Low-Pressure Polymerization of α-Olefins. I. Complex Polymerization (in German), Angew. Chem., **70:** 496-500 (1958).
15. H. Uelzmann, A Theoretical Study on the Mechanism of Ziegler-Type Polymerizations, *J. Polym. Sci.,* **32:** 457-476 (1958).
16. N. A. Adrova, M. M. Koton, and V. A. Klages, Preparation of Organometallic Polymers with Tin Atoms in the Chain, *Vysokomol. Soedin.,* 5(12): 1817-1818 (1963). (*Chem. Abstr.,* **60:** 6936*c*).
17. H. H. Krause, Jr., Organometallic Polymers—A Path to Thermal Stability, *Battelle Tech. Rev.,* **11**(11): 2-6 (1962).
18. *Dow Corning Corp., Block Copolymers of Polysiloxanes and Poly(alkene oxides), Neth. Appl. 6,609,972, Jan. 17, 1967.
19. H. F. Mark and S. H. Atlas, Principles of Polymer Stability, *Polym. Eng. Sci.,* **5:** 204-207 (1965).
20. B. Vollmert, *Grundriss der Makromolekularen Chemie* (Outline of Macromolecular Chemistry), pp. 146, 147, Springer-Verlag, Berlin, 1962. (*Chem. Abstr.,* **58:** 8424*h*).
21. K. Hauffe and C. Seyferth, *Reaktionen in und an festen Stoffen* (Reactions in and on Solids), Springer-Verlag, New York, 1966.
22. J. R. Van Wazer, Inorganic Polymer Chemistry, *J. Macromol. Sci. (Chem.),* **A1**(1): 29-55 (1967).
23. L. H. Van Vlack, *Elements of Materials Science; An Introductory Text for Engineering Students* (2nd ed.), p. 213, Addison-Wesley Publishing Company, Inc., Reading, Mass., 1964.
24. Semiconducting d^8 Metal Complexes Found, *Chem. Eng. News,* **45**(52): 50-51 (Dec. 11, 1967).
25. *L.-H. Lee, Adhesion of High Polymers. I. Influence of Diffusion, Adsorption, and Physical State on Polymer Adhesion, *J. Polym. Sci., Part A-2,* **5:** 751-760 (1967). (*Chem. Abstr.,* **67:** 44168*y*).
26. P. Weiss, The Polymer Concept in Adhesives, *J. Polym. Sci., Part C,* **12:** 169-183 (1966).
27. H. F. Mark, Future Trends for Improvement of Cohesive and Adhesive Strength of Polymers, in *Adhesion and Cohesion,* P. Weiss (Ed.), Symposium Proceedings, Warren, Mich., 1961, pp. 240-269, Elsevier Publishing Company, New York, 1962.
28. S. M. Atlas and H. F. Mark, Possible Research Projects in the Field of Adhesives, *Soc. Plastic Eng. J.,* **20:** 314-315 (1964).
29. S. S. Voyutskii, *Autohesion and Adhesion of High Polymers,* translated from Russian by S. Kaganoff (translation editor V. L. Vakula), Interscience Publishers, New York, 1963.
30. A. F. Lewis and L. J. Forrestal, Chemical Nature of Polymer to Metal Adhesion, in *Adhesion,* ASTM Special Technical Publication No. 360, pp. 59-77, American Society for Testing and Materials, Sixty-sixth Annual Meeting, Atlantic City, 1963, American Society for Testing and Materials, Philadelphia, 1964.
31. D. Taylor, Jr., and J. E. Rutzler, Jr., Adhesion Using

Molecular Models. Adhesion of Polyethylene and Poly-(vinyl Chloride) to Metals, *Ind. Eng. Chem.*, **50**: 928-934 (1958).

32. D. D. Eley (Ed.), *Adhesion*, p. 38, Oxford University Press, London, 1961.
33. J. R. Huntsberger, Mechanisms of Adhesion, *J. Paint Technol.*, 39(507): 199-211 (1967) (*Chem. Abstr.*, **67**: 3358*s*).
34. D. D. Eley (Ed.), *Adhesion*, p. 37, Oxford University Press, London, 1961.
35. *Ibid.*, p. 273.
36. H. Grinsfelder, Adhesion of Organic Surface Coatings, in *Adhesion and Adhesives: Fundamentals and Practice*, Symposium at Case Institute of Technology, Cleveland, Ohio, and Conference of Society of Chemical Industry, London, 1954, pp. 29-33, Society of Chemical Industry, London, and John Wiley & Sons, Inc., New York, 1954 (*Chem. Abstr.*, **49**: 9191*b*).
37. N. A. DeBruyne, The Physics of Adhesion, *J. Sci. Instr.*, 24: 29-35 (1947).
38. R. W. Kaercher and G. W. Blum, Bonding Butyl Rubber to Brass, *Ind. Eng. Chem.*, **43**: 488-493 (1951).
39. S. Buchan and W. D. Rae, Chemical Nature of the Rubber-to-Brass Bond, *Trans. Inst. Rubber Ind.*, **21**: 323-342 (1946) (*Chem. Abstr.*, **40**: 4241_5).
40. *W. G. H. Robins, Rubber–Metal Bonding by the Brass Plating Process, D.I.C. Thesis, University of London, 1946.
41. W. C. Wake, *Rubbers in Adhesion and Adhesives*, Vol. 1, 2nd ed., pp. 375-415, especially 403-404, R. Houwink and G. Salomon (Eds.), Elsevier Publishing Company, New York, 1965.
42. S. Buchan, *Rubber to Metal Bonding*, 2nd ed., Crosby Lockwood & Sons, London, 1959.
43. S. S. Voyutskii and Yu. I. Markin, Adhesion of High Polymers to Metals. I. Adhesion of Various Elastomers to Aluminum and Zinc, *Vysokomol. Soedin.*, **4**: 926-934 (1962) (*Chem. Abstr.*, **59**: 1817*d*).
44. S. S. Voyutskii, Yu. I. Markin, V. M. Gorchakova, and V. E. Gul, How Temperature Dependence and Activation Energy Affect Adhesion of Polymers to Metals, *Adhesive Age*, 8(11): 24-28 (1965) (*Eng. Index*, 1965, p. 9).
45. Y. Tamai and S. Tanaka, Contact Angle Characteristics and Breaking Strength of Paint Plastic Film, *J. Appl. Polym. Sci.*, **11**(2): 297-304 (1967) (*Eng. Index*, 1967, p. 1904).
46. F. E. Bartell and B. R. Ray, Wetting Characteristics of Cellulose Derivatives. I. Contact Angles Formed by Water and Organic Liquids, *J. Amer. Chem. Soc.*, **74**: 778-783 (1952).
47. V. I. Spitsyn, P. I. Zubov, V. Ya. Kabanov, and Z. P. Grozinskaya, Effect of Radiation on Polyethylene Adhesion to Aluminum (in Russian), *Vysokomol. Soedin.*, 8(4): 604-612 (1966), (*Chem. Abstr.*, **65**: 2421*f*). [*Polym. Sci. (USSR)*, 8(4): 662-671 (1966), (*Eng. Index*, 1967, p. 1947)].
48. R. N. Chapman and P. Colombo, A Method for the Improvement of the Bonding Properties of Polyethylene by Gamma Irradiation, USAEC Report BNL-2431, Brookhaven National Laboratory, August 1955 [*Nucleonics*, **13**(10): 13 (1955)].
49. V. B. Kazanskii, G. B. Pariiskii, and V. V. Voevodskii, Use of Electron Paramagnetic Resonance (EPR) for the Study of Hydrogen Atom Properties and Defects Obtained by Irradiation of Silica Gel (in Russian), *Tr. 2-go Vses. Soveshch po Radiats. Khim. Akad. Nauk SSSR, Otd. Khim. Nauk, Moscow*, 1960, pp. 656-659 (pub. 1962) [AEC-tr-6228, pp. 702-706 (1964)].
50. V. B. Kazansky, G. B. Pariisky, and V. V. Voevodsky, Radiation-Induced Processes on the Silica-Gel Surface, *Discussions Faraday Soc.*, **31**: 203-208 (1961).
51. G. B. Pariiskii, Y. A. Mishchenko, and V. B. Kazanskii, On the Nature of Surface Radiation Defects in Irradiated Silica Gel. I. Mechanism of the Adsorption of Hydrogen, *Kinetics Catalysis (USSR)*, **6**: 558-564 (1965).
52. J. H. E. Griffiths, J. Owen, and I. M. Ward, Paramagnetic Resonance in Neutron-Irradiated Diamond and Smoky Quartz, *Nature*, **173**: 439-440 (1954).
53. F. Seitz, The Effects of Irradiation on Metals, *Science*, **138**: 563-571 (1962).
54. V. B. Kazanskii and G. B. Pariiskii, Electron-Spin-Resonance Signal from Ethyl Radicals Adsorbed on Silica Gel, *Kinetics Catalysis (USSR)*, **2**: 460-461 (1961).
55. V. B. Kazanskii, G. B. Pariiskii, I. V. Aleksandrov, and G. M. Zhidomirov, Electron Paramagnetic Resonance Study of Free-Radical Interaction with a Solid Surface (Silica Gel) (in Russian), *Fiz. Tverd. Tela.*, **5**: 649-659 (1963).
56. V. B. Kazanskii and G. B. Pariiskii, The Electron Spin Resonance Study of Free Radicals Adsorbed on Catalysts, *Proceedings of the 3rd International Congress on Catalysis, Amsterdam, 1964*, Vol. 1, pp. 367-378, 1965. (*Chem. Abstr.*, **63**: 12359*g*).
57. R. Worrall and S. H. Pinner, Heterophase Radiation Polymerization of Isobutene and Methyl Methacrylate, *J. Polym. Sci.*, **34**: 229-242 (1959).
58. A. Charlesby, S. H. Pinner, and R. Worrall, Analysis of Radiation-Induced Ionic Polymerization of Isobutene, *Proc. Roy. Soc. (London), Ser. A*, **259**: 386-402 (1960).
59. A. V. Vlasov, P. Ya. Glazunov, Yu. L. Morozov, I. I. Patalakh, L. S. Polak, S. R. Rafikov, and B. L. Tsetlin, Semiconducting Combined Materials Synthesized by Gas-Phase Graft Polymerization by Irradiation, *Dokl. Akad. Nauk SSSR*, **158**(1): 141-142 (1964). (*Chem. Abstr.*, **61**: 15495*b*).
60. L. S. Polak, P. Ya. Glazunov, Yu. L. Morozov, I. I. Patalakh, S. R. Rafikov, and B. L. Tsetlin, Preparation of Mechanically Resistant Semiconducting Materials by Means of Irradiation Grafting (in Russian), in *Radiats. Khim. Polim. Mater. Simp., Moscow*, **1964**: 164-168 (pub. 1966) (*Chem. Abstr.*, **66**: 95731*p*).
61. A. V. Vlasov, N. V. Mikhailov, L. G. Tokareva, S. R. Rafikov, B. L. Tsetlin, and P. Ya. Glazunov, Irradiation-Induced Graft Copolymerization in the Vapor Phase, *Khim. Volokna*, **1963**(6): 24-28 (*Chem. Abstr.*, **60**: 12156*g*).
62. B. L. Tsetlin, S. R. Rafikov, L. I. Plotnikova, and P. Ya. Glazunov, Radiation-Induced Grafting of Polymeric Chains to the Surface of Mineral Particles, *Tr. 2-go Vses. Soveshch po Radiats. Khim. Akad. Nauk SSSR, Otd. Khim. Nauk, Moscow, 1960*, pp. 497-500 (pub. 1962) [AEC-tr-6228, pp. 529-533 (1964)].
63. B. L. Tsetlin, Some Possibilities of Radiation Polymerization from the Gaseous Phase on Solid Base, in *Industrial Uses of Large Radiation Sources*, Conference Proceedings,

Salzburg, 1963, Vol. 1, pp. 289-300, International Atomic Energy Agency, Vienna, 1963 (STI/PUB/75).

64. V. A. Kargin and N. A. Plate, Chemical Grafting on Crystal Surfaces, *Vysokomol. Soedin., Vses. Khim. Obshchestvo im D. I. Mendeleeva*, 1(2): 330-331 (1959). (*Chem. Abstr.*, 53: 19437*c*)
65. E. V. Egorov, P. D. Novikov, D. R. Razgon, and B. L. Tsetlin, Radiation-Chemical Synthesis of New Ion-Exchange Sorbents of Organomineral Nature (in Russian), *Dokl. Akad. Nauk SSSR*, 146: 1360-1362 (1962).
66. Yu. L. Morozov, N. I. Vitushkin, P. Ya. Glazunov, S. R. Rafikov, A. I. Khomutov, and B. L. Tsetlin, Radiation Gas-Phase Graft Copolymerization on Glass Fibers (in Russian), *Radiats. Khim. Polim. Mater. Simp., Moscow, 1964*, pp. 160-164 (pub 1966) (*Chem. Abstr.*, 66: 95732q).
67. V. G. Berezkin, Yu. A. Kolbanovskii, and E. A. Kyazimov, Kinetics of the Radiation Polymerization of Acrylonitrile from the Gas Phase on a Mineral Substrate, *Kinetics Catalysis (USSR)*, 7: 170 (1966).
68. I. A. Uskov, L. I. Tertykh, V. P. Solomko, and Yu. N. Polishchuk, Radiation Polymerization of Methyl Methacrylate and Styrene in Presence of Mineral Fillers, *Polym. Sci. (USSR)*, 8(1): 24-29 (1966). [*Eng. Index*, 1967, p. 1889].
69. M. Magat, Ionic Polymerizations Initiated by Ionizing Radiation (in French), *Macromol. Chem.*, 35: 159-173 (1959).
70. V. B. Kazanskii, G. B. Pariiskii, and V. V. Voevodskii, A Study by the Electron Paramagnetic Resonance Method of the Properties of Hydrogen Atoms on the Surface of Silica Gel, *Kinetics Catalysis (USSR)*, 1: 504-506 (1960).
71. Yu. G. Tarasenko, I. A. Uskov, and V. P. Solomko, Effect of Kaolin on the Properties of Poly(methyl Methacrylate) and on Polystyrene, Ukr. Khim. Zh., 30(1): 86-90 (1964).
72. H. Yasuda and M. F. Refojo, Graft Copolymerization of Vinylpyrrolidone onto Polydimethylsiloxane, *J. Polym. Sci., Part A*, 2: 5093-5098 (1964).
73. T. P. Kornienko, Yu. N. Polishchuk, T. G. Zelenchukova, and M. V. Polyakov, Influence of Mineral Additives on the Radiation Polymerization of Vinyl Monomers (in Russian), *Radiats. Khim. Polim. Mater. Simp., Moscow*, 1964, pp. 69-74 (pub. 1966) (*Chem. Abstr.*, 67: 11918*q*).
74. E. L. Warrick, Method of Preparing Organosilicon Graft Polymers, U. S. Patent No. 2,959,569, Nov. 8, 1960 (filed July 5, 1956) (British Patent 806,582, Dec. 31, 1958).
75. H. G. Ingersoll, Irradiated Refractory Compound as Polymerization Initiator, British Patent No. 866,768, May 3, 1961 (*Chem. Abstr.*, 55: 26538*f*).
76. New Class of Materials Formed by Bonding Clays, *Chem. Eng. News*, 42(43): 70, 72 (Oct. 26, 1964).
77. P. G. Nahin and P. S. Backlund, Organoclay–Polyolefin Compositions, U. S. Patent No. 3,084,177, Apr. 2, 1963 (filed Apr. 4, 1961).
78. P. G. Nahin, Organoclay–Polymer Compositions, U. S. Patent No. 3,248,314, Apr. 26, 1966 (filed Mar. 14, 1960).

Section V

Isotopes and Radiation Technology

Isotopic Heat and Power Development

English Colloquium on Isotopic Thermoelectric Generators

Summarized by F. E. McKinney

Abstract: *The proceedings of the Colloquium on Isotopic Thermoelectric Generators, held in London, Jan. 30, 1968, are summarized. The seven papers cover general principles of radioisotopic thermoelectric generators, the RIPPLE series of generators, properties of thermoelectric materials, thermoelectric conversion theory, civil aviation applications, marine navigation applications, and other practical applications. The full text of the papers is included in* Isotopic Generator Information Centre Newsletter, *No. 6, May 1968.*

New developments in isotopic thermoelectric generators in the United Kingdom were reported at a colloquium held in London on Jan. 30, 1968. The full text of the seven papers presented at the symposium comprised a recent issue[1] of the *Isotopic Generator Information Centre Newsletter. Isotopes and Radiation Technology* reports in this issue[2] on recent developments in Germany, and a past issue[3] covered European developments in some detail. The seven papers presented at the recent English colloquium are summarized here.

General Principles

L. S. Phillips[4] (G. V. Planer Ltd.) reviewed the general principles of radioisotopic generators. A typical generator is made up of a radioisotope heat source, one or more thermoelectric modules to convert heat energy to electrical energy, and a system to convert the normally low d-c output of the modules to a suitable a-c or d-c voltage.

The radioisotopes chosen for the heat source must satisfy several criterions: (1) The half-life should be sufficiently long (the RIPPLE* generators use ^{90}Sr which has a half-life of about 28 years). (2) The specific thermal power (watts/g) should be high; this requirement is difficult to satisfy since specific thermal power is high for radioisotopes with short rather than long half-lives. Increasing power density (watts/cm^3) is one way of increasing specific thermal power. (3) The type of radiation emitted by the radioisotope is important because this determines the amount of shielding needed. The more the shielding, the higher the weight of the unit. (4) The isotope must be available in a chemically stable form and, of course, costs should be kept down by using an inexpensive radioisotope. The eight isotopes that best fit these criterions are ^{60}Co, ^{90}Sr, ^{137}Cs, ^{147}Pm, ^{228}Th, ^{232}U, ^{238}Pu, and ^{244}Cm.

The conversion efficiency of a thermoelectric generator is given by

$$\eta = \frac{T_h - T_c}{T_h} \cdot \frac{\sqrt{1+ZT}-1}{\sqrt{1+ZT}+(T_c/T_h)}$$

where Z is termed the thermoelectric *figure of merit* and is given by $Z = (\alpha^2\sigma)/K$

and α = mean Seebeck coefficient† for thermocouple materials
σ = mean electrical conductivity of materials
K = mean thermal conductivity of materials
T_h = temperature of hot junction
T_c = temperature of cold junction

*RadioIsotope Powered Prolonged Life Equipment.

†The Seebeck coefficient is a temperature-dependent measurement in μv/deg of the amount of current generated by the Seebeck effect.—*The Editor*

For a high Z value, a material must have a high Seebeck coefficient, high electrical conductivity, and low thermal conductivity. Thus Z is temperature dependent, and thermoelectric materials are usually suitable only over certain temperature ranges. From 0 to 300°C, materials based on Bi_2Te_3 are best; the upper operating limit is set by the decrease in Seebeck coefficient and increase in thermal conductivity with rising temperature. From 300 to 500°C the best materials are PbTe, GeTe, $AgSbTe_2$, SnTe, and alloys of these compounds; the upper operating limit in this group is defined by chemical stability. Above 500°C, materials based on germanium–silicon alloys and iron disilicide are used; the upper limit on isotopic sources imposed by material technology is about 650°C. Theoretical conversion efficiencies are 4 to 5% for the 0 to 300°C group, 9 to 10% for the 300 to 500°C materials, and 2 to 3% for the last group.

Thermocouple pairs are usually connected in series electrically and in parallel thermally to provide a high, usable output. For maximum reliability the metal strap connecting the p- and n-type thermoelectric alloys should be compatible both metallurgically and electrically, yield minimum electrical contact resistance, and have a low diffusivity into the alloys. There must be an electrically insulating layer between the faces of the module and the heat sink. This layer must not cause too large a thermal drop; about 10°C is acceptable.

Stringent control during manufacture is very necessary. The 4-watt RIPPLE-IX contains 8 modules with a total of 1600 contacts; none must fail. Various designs incorporating redundancy are under assessment to alleviate the problem.

Because thermoelectric generators are low-voltage, high-current devices, the output must usually be converted to a suitable voltage and current level by a power conditioning unit. If the power is used to drive a pulsed system, the output can be used to trickle-charge a storage battery. If the power is used to drive a continuous system, the power must be converted to a highly stable type. In any case, the conditioning unit should be as efficient as possible; efficiencies greater than 80% have been obtained with d-c to d-c inverters having an output greater than 3 volts.

For safety, a shield must surround the radioisotope generator. In addition to being readily available and inexpensive, the shielding material should have the following properties: a strong attenuation of beta and gamma radiation, ease of fabrication, stability, high melting point, and compatibility with the other generator materials at high temperatures. The last two requirements are dictated by the necessity to preserve and contain the radioactive source in case of accidental fire. The usual shielding materials are depleted uranium, heavy-metal alloys, lead, steel, and concrete. The RIPPLE generators are shielded with depleted uranium.

To obtain maximum conversion efficiency, all the heat produced by the source must pass through the thermoelectric modules surrounding the source. The heat insulating material must have a very low thermal conductivity and a high operating temperature. The later RIPPLE generators use a new material called Microtherm; it is a mixture of microporous silica (filled with an inert gas), a fibrous bonding material, and an opacifier to reduce the infrared transparency. Because the pore size of the silica is less than the mean free path of the gas molecules, the normal conduction process by means of kinetic molecular collisions does not occur and heat is transferred only by the accommodation between the gas molecule and the cavity wall. Overall thermal conductivities as low as 2×10^{-3} watts/(cm)(°C) in argon have been obtained in the RIPPLE generators.

RIPPLE generators have a 5% efficiency of conversion of heat to electric power, a 15 to 20% drop in output because of radioactive decay over the 5-year life of the generator, a voltage-converter efficiency of about 80%, and a battery storage efficiency of 85%. These result in an overall efficiency of about 2%. The next series of RIPPLE generators is expected to have an overall efficiency of between 6 and 7%.

RIPPLE Applications

F. W. Yeats[5] (AERE, Harwell) briefly reviewed the RIPPLE series of generators. Power and efficiency data on the generators built or under construction are given in Table 1. The first two RIPPLE's were built for demonstration purposes, but they are still operating. An experimental prototype, RIPPLE-III, was built next

Table 1 Power and Efficiency of RIPPLE Generators

RIPPLE No.	Thermal power of isotope, watts	Generator power (output at 20°C), watts	Generator efficiency, %
I	4.40	0.075	1.705
II	4.40	0.092	2.091
III	29.05	0.795	2.737
V	47.60	1.490	3.130
VI	59.65	2.000	3.353
VII	51.55	1.545	2.997
IX	199.50	6.400	3.275

and is on loan to the U. S. Navy who will use it to power an acoustic beacon almost 7000 ft deep in the Pacific Ocean. Models V, VI, and VII are being used to power navigational lights (Figs. 1 to 3) off the Kent coast, in Sweden, and in Denmark. Two other RIPPLE generators have been built for special purposes: RIPPLE-IV will power a submerged transistorized telecommunication repeater, and RIPPLE-IX is powering an aircraft radio beacon.*

The overall system efficiency—the most important of the many types of efficiencies quoted in the literature on radioisotopic generators—is defined as the ratio of the electric power at the end of the generator's lifetime to the thermal power at the beginning of the generator's lifetime. The kilowatt-hours stored per unit weight and volume of the RIPPLE generators are impressive when compared to other small power sources (Table 2).

Power output of an isotopic generator is reduced as the radioisotope decays; power output also varies with the ambient temperature. To compensate for these fluctuations, the designer of a generator must arrange for the power output at the end of the lifetime of the unit under the worst ambient temperature to be the maximum power ever required for the system powered by the unit.

The power conditioning unit of a radioisotopic thermoelectric generator must provide a suitable voltage conversion and regulate the fluctuations of generator voltage caused by radioisotope decay and ambient-temperature variation. The three generators providing power for navigational lights produce approximately 4 volts; a transistorized d-c to d-c unit converts this to 12 volts, which is fed to Ni–Cd batteries. Conversion efficiency is 80% with the batteries half-charged.

*Fully described in a paper that is reviewed later in this article.

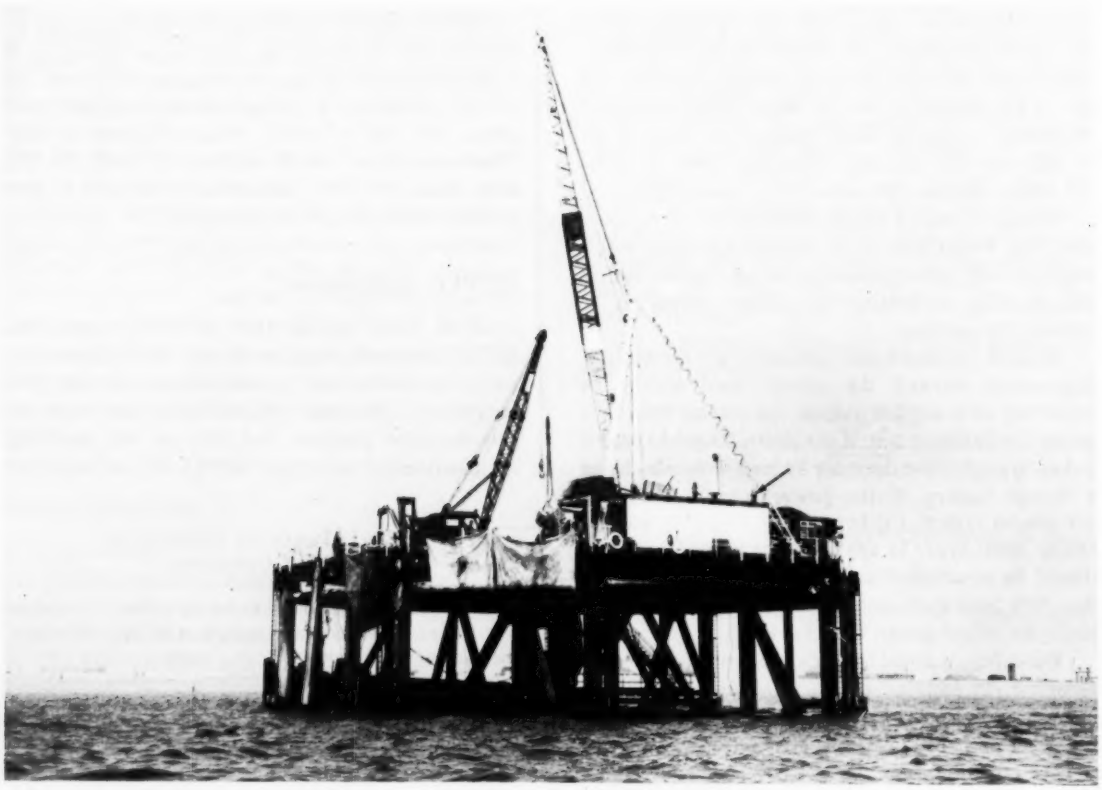


Fig. 1 Cooling-water intake platform off Dungeness, England, marked by light powered by RIPPLE-V (arrow). Photograph by the courtesy of F. W. Yeats, AERE, Harwell.

RIPPLE-IX produces 8 volts; its power conditioning unit has been designed to convert the output to the 65 volts needed to operate the aircraft beacon.

Reliability is one of the advantages emphasized for isotopic generators. They have no moving parts; they contain their fuel supply for the required lifetime; and they possess a fuel system that is virtually indestructible. The only vulnerable part of an isotopic generator is the thermoelectric module, which contains many electrical contacts. To date, no RIPPLE generators have failed in service, and they have logged a total of over 82,000 hr, with RIPPLE-I running 25,300 hr so far.

There are three improvements being emphasized in the RIPPLE-X generators, the next series of radioisotopic thermoelectric generators. The present thermoelectric material can be improved; further improvements are possible by cascading different materials—either in the same module or by joining two modules to allow a higher source temperature and, thus, a higher Carnot efficiency. Heat losses can be minimized by careful design and selection of thermal insulation. The power conditioning unit can be made more efficient, too. Some measure of standardization and simplification is necessary to keep construction costs low.

Fig. 2 Navigational light in Stockholm Archipelago powered by RIPPLE-VI. Photograph by the courtesy of F. W. Yeats, AERE, Harwell.

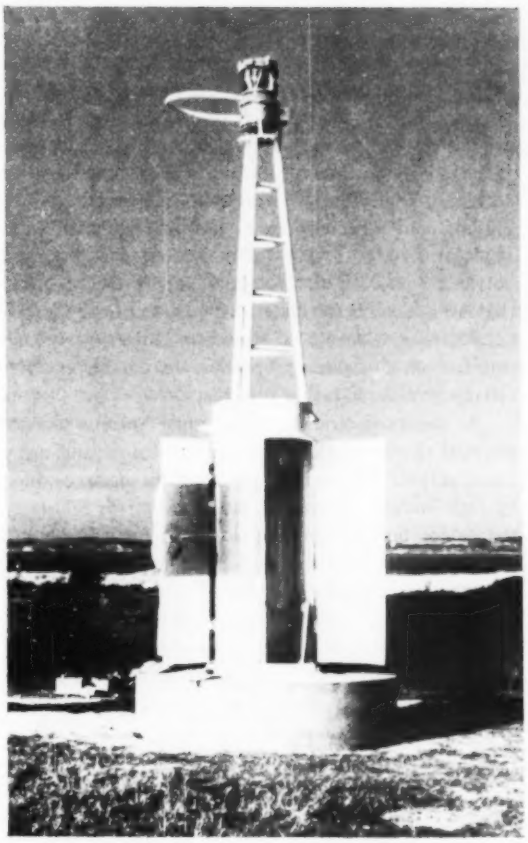


Fig. 3 Navigational light at Zealand, on the coast of Denmark, powered by RIPPLE-VII. Photograph by the courtesy of F. W. Yeats, AERE, Harwell.

Table 2 Weight and Volume Values for Different Power Sources

Power source	Weight, kw-hr/lb	Volume, kw-hr/cu ft
Primary cells	0.10	8
Secondary cells		
Lead acid	0.13	1.9
Nickel–cadmium	0.07	0.7
Fuel cells	0.1 to 0.2	6 to 15
RIPPLE generators	2.5 to 3.2	100 to 150

Properties of Thermocouple Materials

A detailed report on the thermoelectric and physical properties of thermocouple materials and on the semiconductors successfully developed for working generators was presented by Christie[6] (G. V. Planer Ltd.). The selection of materials is governed by the efficiency equation, presented in the earlier talk by Phillips, and especially by Z, the figure of merit. The numerator, $\alpha^2\sigma$, reaches a maximum nearly always between 3×10^{18} and $3 \times 10^{19}/cm^3$. Increases above this level decrease the electronic portion of the thermal conductivity. Because of the importance of temperature for some of these properties, the parameter ZT is usually considered rather than just Z.

A thermoelectric material must possess certain physical characteristics to be suitable for module-array construction. The melting point of the material must be high enough to prevent deterioration or failure of the device but low enough to permit preparation and handling of the material; in practice the upper working temperature is usually set at about three-fourths of the melting point of the material. The volatility of the compound should be as low as possible at the designed operating temperature; vapor pressures above 0.07 mm Hg can sometimes cause reduction in performance. Preferential diffusion of a constituent along the thermal gradient is undesirable. In general, this can be avoided by operating at temperatures well below the melting point, since diffusion becomes a serious problem only at about 80 to 90% of the melting point of the material (Tamman's Rule). In addition, brittleness, shear strength, and thermal expansion must also be considered.

Thermocouples are constructed of alternate *p*- and *n*-type limbs connected by a strap of suitable contacting materials. Since doping semiconductors to form *p* or *n* material is relatively easy, a large number of semiconductors have been examined for use in thermocouples. Some of the most promising are listed in Table 3 with their figures of merit.

Table 3 Figure of Merit, Z, for Thermoelectric Materials

Material	Temperature, °K	Figure of merit, deg^{-1}
n-type Bi_2Te_3	300	2.00×10^{-3}
(75% Bi_2Te_3,	375	2.80×10^{-3}
25% Bi_2Se_3)	500	2.13×10^{-3}
	600	2.13×10^{-3}
p-type Bi_2Te_3	350	3.12×10^{-3}
(75% Sb_2Te_3,	450	2.0×10^{-3}
25% Bi_2Te_3)	550	1.1×10^{-3}
p-type PbTe	450	0.61×10^{-3}
(0.25% Bi)	520	0.85×10^{-3}
	620	1.00×10^{-3}
	800	1.00×10^{-3}
	890	0.98×10^{-3}
n-type $GeTe/AgSbTe_2$	320	0.60×10^{-3}
	520	1.20×10^{-3}
	670	1.95×10^{-3}
	770	2.20×10^{-3}
	870	1.97×10^{-3}
n-type Si–Ge alloy	500	1.7×10^{-4}
	800	6.6×10^{-4}
	900	7.2×10^{-4}
	1000	7.5×10^{-4}
	1100	7.25×10^{-4}
p-type Si–Ge alloy	300	1.7×10^{-4}
	500	4.0×10^{-4}
	750	5.3×10^{-4}
	1000	6.5×10^{-4}
	1100	6.4×10^{-4}
p-type $FeSi_2$	500	2.4×10^{-4}
(5% $CoSi_2$)	700	2.1×10^{-4}
	900	1.4×10^{-4}
n-type $FeSi_2$	500	2.7×10^{-4}
(2% $AlSi_2$)	700	2.9×10^{-4}
	900	2.1×10^{-4}

Thermoelectric Materials in Use

Bismuth telluride alloys, the most important thermoelectric materials in use today, are often mixed with Sb_2Te_3 and Bi_2Se_3. The loss of volatile tellurium from the material is the biggest production problem. This loss has been reduced by melting and mixing the components, in vacuum, in silica ampules. After cooling, the material is ground, pressed, and then sintered at a relatively low temperature. The material can exhibit anisotropy because of crystal orientation during pressing; in this case the material must be carefully cut along the desired plane. Recently, isostatically pressed material has not exhibited anisotropy. Another production problem is the fabrication of the strap contacting the *p*- and *n*-type limbs; the two difficulties are to achieve a low contact resistance for maximum output and to prevent diffusion at operating temperature. Copperplated strap is usually used, with a protective layer of nickel plate to prevent diffusion.

The serious limitation of bismuth telluride alloys is their upper operating limit. At 200°C the figure of merit starts decreasing, and above 300°C their efficiency is poor. These alloys are reliable, however, and every generator in the RIPPLE series uses them.

Lead telluride alloys offer greater efficiencies than do bismuth telluride alloys because of their higher operating temperatures. Many methods of preparing the properly doped alloys have been reported; G. V. Planer Ltd.[7] has obtained good results by hot pressing a previously chill-cast material at 700°C and 7000 psi. Production of *n*-type lead telluride gives material that is thermally stable, but *p*-type material often has problems with dopant diffusion. Alternative telluride-based alloys— $AgSbTe_2$ and $GeTe/AgSbTe_2$—have been tried with good results.

The thermoelectric materials discussed so far require the use of expensive, high-purity starting materials; hence great care is necessary during manufacture. Another difficulty is making satisfactory contacts. The great advantage of telluride alloys is their relatively high efficiencies at low temperatures; however, the volatility of tellurium puts an upper limit of 500 to 600°C on their use.

Silicon-based alloys show promise for use at temperatures up to at least 1000°C. Polyphase ingots of GeSi using phosphorus and boron as dopants are converted into a single-phase alloy by milling to a fine powder then hot pressing, using a carbon die. Although the material thus prepared contains large quantities of impurities, which affect the physical properties of the material, the thermoelectric properties are similar to those of carefully prepared, zone-leveled material. The figure of merit for these materials is lower than that of the telluride-based alloys, but their temperature range is wider and they are less volatile.

Despite a low figure of merit, iron disilicide materials offer the advantages of low cost and ease of fabrication. A large temperature difference is required to obtain reasonable performance from these materials; iron disilicide alloys have been successfully used in flame-powered generators operating at a hot sink temperature of 800°C and a temperature difference of 600°C.

Thermoelectric Conversion Theory

H. J. Goldsmid[8] (Bath University of Technology) reported on the theoretical aspects of thermoelectric materials. For the past 50 years, the efficiency of thermocouples has been expressed[9] as a figure of merit*

$$Z = \frac{(\alpha_p - \alpha_n)^2}{KR}$$

*This is the mathematically exact expression, whereas the expression used by Phillips and Christie earlier is an approximation.—*The Editor*

where α = absolute Seebeck coefficient
K = total thermal conductance of the thermocouple
R = electrical resistance
p refers to positive branch
n refers to negative branch

The figure of merit is highest when the product KR is at its minimum value; this occurs when $R_p/K_p = R_n/K_n$. In the relationships that give the performance of a thermoelectric energy converter, the figure of merit always appears as a product with the average absolute temperature; this is one reason the dimensionless figure of merit, ZT, is adopted. If t is the ratio of sink to source temperature, the efficiency of generation η is given by

$$\eta = (1 - t)\left(\frac{M - 1}{M + t}\right)$$

where $M = \sqrt{1 + ZT}$. Table 4 gives the values of η for various values of ZT, M, and t.

Table 4 Generation Efficiency η for Selected Values of the Figure of Merit ZT and the Temperature Ratio t

		Efficiency of Generation, η			
ZT	M*	$t = 0.75$	$t = 0.5$	$t = 0.25$	$t = 0$
0.25	1.118	0.016	0.036	0.065	0.106
0.5	1.225	0.028	0.065	0.114	0.183
1	*1.411*	*0.048*	*0.108*	*0.186*	*0.291*
2	1.732	0.074	0.164	0.277	0.422
4	2.236	0.103	0.226	0.372	0.552
8	3.000	0.133	0.286	0.462	0.667
∞	∞	0.25	0.5	0.75	1

*$M = \sqrt{1 + ZT}$.

When $ZT = 1$, the efficiency of generation is about 20% that of an ideal heat engine for a temperature ratio of 0.25; this efficiency rises to nearly 30% of the ideal value for larger temperature differences between the source and the sink.

When Altenkirch[9] studied thermoelectric energy conversion, only metals and alloys were available. For such materials the ratio of thermal to electrical conductivity is given by $K/\sigma = 2.44 \times 10^{-8}\ T$. However, even the best alloys—such as constantan and Chromel—have ZT values less than 0.1; obviously the efficiency of metallic thermocouples is very low. The Seebeck coefficients of semiconductors are much

greater than those of metals—so much so, in fact, that if the Wiedemann–Franz* law were obeyed, ZT would be about 40. Semiconductors do not obey this rule, however. In semiconductors, heat transport is primarily through lattice vibrations and not by electrons.

The Seebeck coefficients in practical thermoelectric devices are always about ±200 μv/deg. The properties of a typical semiconductor (Fig. 4) vary

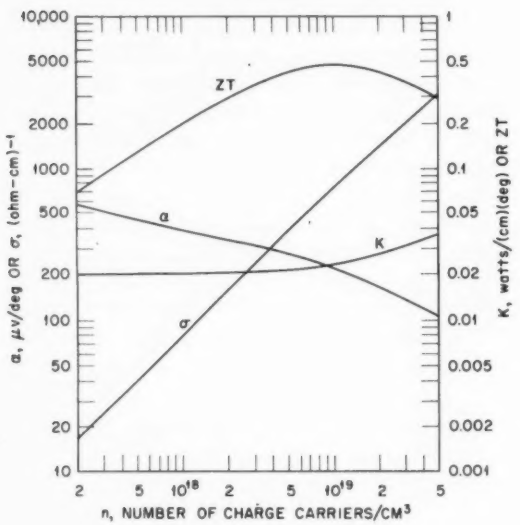


Fig. 4 Variation of semiconductor properties with concentration of charge carriers. ZT = dimensionless figure of merit; K = thermal conductivity; σ = electrical conductivity; α = mean Seebeck coefficient.

with concentration of charge carriers, which is adjustable by doping or purifying the material. The Seebeck coefficient is proportional to the total energy, both kinetic and potential, transported by the charge carriers. The total energy in turn depends on the position of the Fermi level; the number of charge carriers also depends on the position of the Fermi level. Carrier concentration n increases as the energy of the carriers decreases. However, the electrical conductivity varies linearly with the carrier concentration. The optimum carrier concentration, usually around $10^{19}/cm^3$, depends strongly on the density of states for the charge carriers as expressed in terms of the ratio of effective mass m^* to the mass m_0 of a free electron. For a semiconductor such as indium antimonide, the optimum carrier concentration might be $10^{16}/cm^3$ since its electrons have a small effective mass. In any case, the optimum Seebeck coefficient remains fairly close to ±200 μv/deg.

The presence of both electrons and vacancies—negative and positive charge carriers—is undesirable, since the thermoelectric effects of one cancel those of the other. Another reason is that the thermal conductivity is large for low electrical conductivities because, in a mixed conduction region, both carriers transport ionization energy along a temperature gradient. Some materials that would otherwise be good for thermoelectric applications have both electrons and vacancies present at relatively low temperatures. Mixed conduction is a serious problem in bismuth telluride at elevated temperatures, and lead telluride has a better figure of merit when the temperature exceeds 200°C.

The semiconductors most widely used in thermoelectric energy conversion are based on tellurium compounds with Group IV or Group V elements. However, electrical properties of compounds depend on departures from stoichiometric formulas; tellurides are notorious for stoichiometric deviations. They tend to dissociate at only moderately high temperatures. Tellurium itself is volatile, and, even if not very poisonous, it does produce unpleasant symptoms in the body. Why then are tellurides used?

An extrinsic semiconductor with Seebeck coefficient of about ±200 μv/deg is desired, with the highest possible ratio of electrical conductivity to thermal conductivity. The electrical conductivity, as explained above, depends on the effective mass ratio m^*/m_0; the thermal conductivity will be lowest when the lattice component K_L is smallest. These considerations lead to the conclusion that the semiconductor should have a high value for the factor $(\mu/K_L)\,(m^*/m_0)^{1.5}$, where μ is carrier mobility; this factor is greatest for compound semiconductors of high mean atomic weight, namely, tellurides of the heavier metals.

The best thermoelectric materials are not simple compounds but disordered solid solutions (or alloys) involving compounds or elements, because disorder—through effective scattering of lattice waves—lowers the lattice thermal conductivity. Disorder has little effect on carrier mobility since the short-range disorder in solid solutions is ineffective in scattering the large wavelengths in a semiconductor material. Thus thermocouples for use at ordinary temperatures are made of Bi_2Te_3–Sb_2Te_3 alloy in the p-type branches and of Bi_2Te_3–Bi_2Se_3 alloy in the n-type branches.

*The Wiedemann–Franz law states that the ratio of thermal to electrical conductivity is independent of electron mean free path and electron velocity and proportional to the temperature.—*The Editor*

Obviously, the existing thermoelectric materials have been selected after much careful theoretical and experimental work. How much improvement can be expected? If the best product—$\mu(m^*/m_0)^{1.5}$— for any known material were combined with the lowest possible lattice thermal conductivity, ZT would be about 4. More realistically, the practical limit of ZT is about 2. Thus research may improve semiconductor compounds but will never make thermocouples into really efficient heat engines. There are, however, some straightforward ways to improve present-day thermoelements. For bismuth telluride these improvements are better alignment of crystal planes, greater homogeneity of the material, and more uniform distribution of doping material.

Civil Aviation Application

The potential use of isotopic generators and the actual use of one generator in civil aviation was described by Clark[10] (BOAC). Civil aviation today is a large, dynamic business involving national administration, equipment manufacturers, and airline operators. About 6 thousand aircraft flew 4.5 thousand million kilometers in 1966 producing a gross income of about $11 thousand million. Some possible applications of thermoelectric generators were reviewed at the 1966 Harwell conference,[11] and Clark repeated them briefly. There are three specific areas of application: power supplies for aircraft, airborne communication and navigation aids, and ground-based communication and navigation aids. On both economic and technical grounds, there are no foreseeable uses for isotopic power supplies for the generation of primary, secondary, or ground support power for aircraft. There are two possible applications of isotopic power supplies for airborne aids: to provide continuous power for highly complex inertial navigation devices and other digital navigation computers and to replace conventional batteries in low-power search and rescue beacons and recorders. Neither application is economically attractive now.

Provision and operation of ground-based communication and navigation aids are the functions of the International Civil Aviation Organization (ICAO) and its 115 national administration members. The ICAO sets standards and provides planning, and the national administrations finance and maintain the equipment. The aids are of six types: very-high-frequency (VHF) communications, long-range communications, airfield approach and landing aids, air traffic control systems, short-range navigation aids, and long-range navigation aids. Power-generation requirements limit possible thermoelectric generator applications to VHF communications, short-range navigational aids, and airfield approach and landing aids.

After the Harwell symposium,[12] discussions between the AERE and the Civil Aviation Department led to selection of a site and a trial application of a radioisotopic thermoelectric generator to power a short-range navigational aid. The site chosen was in the Outer Hebrides islands, off the northwest coast of Scotland. RIPPLE-IX was designed for this application. The unit provides power for a new solid-state fan marker beacon that marks the high ground in the approach to Benbecula airport. The beacon is mounted at almost sea level; it is exposed to the full fury of the North Atlantic and there is only 1 month a year when the temperature does not go below freezing. Furthermore, it can be reached only by traveling 18 hazardous miles along a narrow road and causeway. Power has been provided by two diesel generators, with one or the other operating continuously.

The heat source for RIPPLE-IX is 32 kilocuries of ^{90}Sr in the titanate form. Thermoelectric conversion is achieved by a matrix of p- and n-type Bi_2Te_3 thermocouples mounted on two copper stalks inserted from the ends of the generator. Each copper stalk has four 50-thermocouple elements connected in series and generating 2 volts across a 5-ohm load. The eight elements are connected as four parallel pairs in series, giving about 8 volts (6.4 watts) at 15°C at the beginning of operation. This is converted, d-c to d-c, to the required 65 volts for the fan marker beacon.

Aircraft approaching Benbecula fly at a heading of 244° and an altitude of 1700 ft, a sufficient height to clear the high ground around the airport. When an aircraft passes over the beacon powered by the radioisotopic thermoelectric generator, an indicator on the flight deck of the aircraft lights up, and the pilot knows that descent to the field can begin safely. This first known operational application of radioisotopic power supply to civil aviation hopefully signals further improvement of the safety of civil aviation.

Marine Navigation Application

I. C. Clingan[13] (Trinity House) summarized the use of thermoelectric generators to power marine navigational aids. There are many kinds of marine navigational aids; the ones of interest are the self-contained short-range active aids, i.e., those that emit some signal, with low power requirements. Such devices are floating buoys or isolated beacons; for these aids, fuel cost

itself is usually not high, but transporting the fuel to the aid is often quite difficult and costly. The advantage of the radioisotope generator is its ability to provide power over a 5- or 10-year period; this advantage should not be overestimated, however, because some gas-operated equipment can operate up to 3 years without refueling. Full advantage cannot be taken of the thermoelectric power supply if the equipment it powers is not equally reliable. If maintenance visits can be reduced to quick inspections or simple replacement of electronic parts, the type of craft used to carry out this routine could be smaller and faster, with a further cost reduction.

Trinity House is testing RIPPLE-V on the offshore cooling-water intake platform for the nuclear generating plant at Dungeness. The generator has an output of 1 watt and powers a xenon flashtube which drains 800 mw from a 55 amp-hr 12-volt battery that is charged by the generator. This installation operated satisfactorily until the xenon flashtube failed.

A typical unattended lamp of 200 candela* intensity has an operating cost of 0.4 pence/watt-hr for gas and 1.2 pence/watt-hr for primary cells. An estimate of the costs for a radioisotopic thermoelectric generator is difficult. The costs of the primary cell and basic fuel provide a starting point, and these costs must be balanced against the high cost of transporting other fuel.

Other Practical Applications

J. E. Rice[14] (Submarine Cables Ltd.) described a commercially available isotopic generator (ITG) developed from the RIPPLE series and reviewed the practical applications of radioisotopic thermoelectric generators. Presently available ITG's range in power from a few milliwatts up to 25 watts, with lifetimes of up to 10 years. One such generator, the ITG 109, has been designed by Submarine Cables Ltd. and is commercially available. This generator will provide 5 watts for 5 years; it uses about 20% less ^{90}Sr than the RIPPLE-IX, although its power output is equal. The ITG 109 can operate on land or in the sea; for submarine application, a pressure housing is necessary.

The nominal power of an isotopic generator is the power output at the end of a certain period of time; to say the ITG 109 has a power output of 5 watts for 5 years means that after 5 years' operation, the output will be no less than 5 watts. The period during which the generator provides at least its rated power is called the rated power period (RPP). When a constant power supply is required in excess of the rated voltage of the generator, a unit is used to convert the output to the required voltage.

The cost of fuel is a large part of an isotopic generator's price. Cost per watt decreases as generator size increases; increasing conversion efficiencies and improved heat insulation will also decrease costs. The ITG 109 can power a 50-watt flashing light for 18 hr/day at a cost of 0.5 pence/watt-hr. A 25-watt generator could power a 300-watt light for less than 7 pence/watt-hr.

The Dungeness marine navigational aid has already been described. Two other such flashing lights have been installed: one in the Stockholm Archipelago and the other on the west coast of Zealand, Denmark. The Swedish light is a 10-watt device that flashes 0.3 sec on, 1 sec off, 0.3 sec on, and 4.4 sec off; it operates about 17 hr/day and is switched on and off photoelectrically. The thermoelectric generator trickle-charges two banks of Ni–Cd batteries after the output is converted from 4 volts to 12 volts. Installation, from the time the generator left the dock, took 2 days, including the 25-mile sea journey to the site. The Danish generator was installed in 1 day.

Locations that economically favor use of thermoelectric generators are places where the environment is hostile (e.g., underwater) and places where topography is rugged. The first includes marine navigational lights and other applications such as undersea sonic beacons to mark static sites or provide navigational data; power for remotely located weather-monitoring buoys that are part of the weather satellite system would also fit in this category. Another important underwater application would be power for research and development programs, i.e., data-gathering stations and life-support systems. Remote radiocommunications sites are prime examples of the cases where topography, often together with environment, makes access difficult. VHF and UHF mobile transmitting stations (50-watt requirement), single-circuit radiotelephone links such as those in the Australian outback (10-watt requirement), and microwave radio-relay-system repeaters (two units, 100-watt requirement each) can be powered now or in the near future by radioisotopic thermoelectric generators. Operation at remote jungle sites would be less costly if roads did not have to be built to them for frequent servicing. The same argument holds true for remote sites in northern countries

*A candela is one-sixtieth of the luminous intensity of 1 cm^2 of a blackbody surface at the solidification temperature of platinum; solidification temperature is equal to or lower than the melting point (1769.3°C for platinum).—*The Editor*

where ice and snow limit access; for severe cold conditions, excess thermal heat from the generator can be used to keep temperature-sensitive equipment operative. This form of power generation is essential because of the trend toward more remote and inaccessible electronic installations for civil and military use, where long and trouble-free life without maintenance is vital.

References

1. *Isotopic Generator Information Centre Newsletter*, No. 6, May 1968.
2. G. Rosner, Isotopic Generators in the Federal Republic of Germany, *Isotop. Radiat. Technol.*, **6(2)**: 197-199 (Winter 1968–1969).
3. J. G. Morse, Isotopic Power in Europe—II, *Isotop. Radiat. Technol.*, **6(1)**: 100-108 (Fall 1968).
4. L. S. Phillips, General Principles of Isotopic Thermoelectric Generators, *Isotopic Generator Information Centre Newsletter*, No. 6, pp. 7-17, May 1968.
5. F. W. Yeats, RIPPLE Generators, *Isotopic Generator Information Centre Newsletter*, No. 6, pp. 19-33, May 1968.
6. I. R. A. Christie, Thermoelectric Materials, *Isotopic Generator Information Centre Newsletter*, No. 6, pp.35-46, May 1968.
7. L. S. Phillips, I. R. A. Christie, and G. V. Planer, Development of Nuclear Conversion Thermoelectric Power Generators, in *Industrial Applications for Isotopic Power Generators*, International Symposium, Harwell, England, September 1966, pp. 339-347, Organization for Economic Cooperation and Development, January 1967.
8. H. J. Goldsmid, Theoretical Aspects of Thermoelectric Materials, *Isotopic Generator Information Centre Newsletter*, No. 6, pp. 47-56, May 1968.
9. E. Altenkirch, Uber den Nutzeffekt der Thermosäule, *Physik. Z.*, **10(16)**: 560-568 (Aug. 15, 1909).
10. J. O. Clark, The Economic and Commercial Potential of Isotopic Generators in Civil Aviation, *Isotopic Generator Information Centre Newsletter*, No. 6, pp. 57-73, May 1968.
11. J. O. Clark, S. D. Whiddett, R. C. Meadows, and R. A. Young, Prospects for the Use of Isotopic Power Sources in Civil Aviation Communications and Navigation Systems, in *Industrial Applications for Isotopic Power Generators*, International Symposium, Harwell, England, September 1966, pp. 123-149, Organization for Economic Cooperation and Development, January 1967.
12. J. P. Anchisi (Ed.), *Industrial Applications for Isotopic Power Generators*, International Symposium, Harwell, England, September 1966, Organization for Economic Cooperation and Development, January 1967.
13. I. C. Clingan, Isotopic Generators in Marine Navigational Aids, *Isotopic Generator Information Centre Newsletter*, No. 6, pp. 75-79, May 1968.
14. J. E. Rice, Practical Aspects and Potentialities of Isotopic Generators, *Isotopic Generator Information Centre Newsletter*, No. 6, pp. 80-92, May 1968.

Isotopic Generators in the Federal Republic of Germany*

By G. Rosner†

In the Federal Republic of Germany, efforts in the field of isotopic generators for both space and terrestrial applications are concentrated on systems for transforming the heat produced by an isotopic source into an electric current. These conversion systems—thermionic, thermoelectric, photoelectric, and thermophotovoltaic—have thus far been operated with electric heat, simulating an isotopic heat source to be fabricated later. Some work has also been done on cardiac pacemakers.

Thermionic Conversion

A prototype of a conversion system based on cesium diodes and intended for use in an isotopic generator was set up in October 1967 at Brown, Boveri & Cie. in Mannheim. The electrically heated device is now undergoing lifetime tests. The overall efficiency is estimated to be about 10%. The device is to be fueled with spherical $^{227}Ac_2O_3$ particles coated with ThO_2 and embedded in a tungsten or

*Reprinted, by permission, with minor editorial changes from *Isotopic Generator Information Centre Newsletter*, No. 5, March 1968, pp. 9-12 (ENEA, Paris; CEA–ENEA, Saclay). The German company names mentioned are given as they were in the original article.

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tungsten–rhenium-alloy matrix. Details on the conversion system and heat source are:

Thermionic Conversion System

Emitter temperature, °C	1527 to 1627
Output voltage, volts	0.6 to 0.8
Output current density, amp/cm^2	8 to 10
Electrode efficiency, %	12 to 15
Thermal input power density, watts/cm^2	40 to 60
Generator efficiency, %	10 to 12
Electrode distance, mm	0.2

Fuel

Diameter of Ac_2O_3 particles, μm	200 to 500
Thickness of ThO_2 coating, μm	10 to 25
Volume density of refractory-metal matrix, % of theoretical	25 to 50
Power density, watts/cm^3	46
Wall thickness of refractory-metal capsule, mm	3
Maximum fuel temperature, °C	1860
Thermal power of heat source, watts	250

The complete isotopic generator will weigh about 350 g, and the whole device, including shielding, will weigh only about 1 kg. Thus it will be particularly suitable for space application.

Thermoelectric Conversion

The problems arising in the use of Ge–Si thermoelements seem to be solved. Such thermoelements can be made which are resistant to temperatures of at least 1000°C, especially with regard to contact and bridge material. They can also be made in different shapes—round, square, or rectangular. The resistance of both *n*- and *p*-type elements to radiation for doses up to 2.6×10^8 rads and at temperatures up to 700°C has been tested, using ^{60}Co gamma radiation to simulate ^{90}Sr bremsstrahlung; this corresponds to a generator lifetime of several years. The measured values of α (μv/°C) and ρ (milliohms/cm) before and after irradiation were found to be identical within the limits of the measurement error. These limits are now ±7% but are expected to be decreased.

The Ge–Si thermoelectric conversion systems are being developed by both AEG and Siemens. A thermoelectric conversion system with an output of 50 watts(e), consisting of six modules of twelve thermocouples each, has operated for 2000 hr in air and in vacuum. Before the complete device was set up, the modules and their elements were tested separately for times of up to several years at constant and variable temperatures. Data collected with the complete device were used in optimizing the heat flux. This was done by using longer thermoelements and by transporting the heat from the hot end of the thermoelements by radiation instead of by direct contact. In cooperation with a European nuclear research center, a second isotopic generator, to provide 5 watts(e), is now being developed. Both these devices are for space use. A third system, of 25 watts(e), is being studied by AEG for terrestrial purposes.

A Ge–Si conversion system of 125 watts(e) has been in operation for 1000 hr, and a second model is being made. These "Isolde" models are designed for space use. For terrestrial uses, where residual heat may be carried off by convection, a promising conversion system, called "Tristan," is under construction. This works on a tandem principle: the cold end of the primary Ge–Si thermoelement is in contact with the hot end of a secondary Bi_2Te_3 element, and the cold end of this Bi–Te thermoelement is attached to a cooling cylinder of copper metal. Twenty-four of these thermoelements surround the heat source, and the heat is transferred to them by radiation. The planned temperature difference between the heat source and the last cold end is ~800°C, with the following differences between the hot and cold ends:

Ge–Si element: hot end 900 to 930°C, cold end 300°C

Bi_2Te_3 element: hot end 250°C, cold end 60 to 100°C

This system is to use ^{90}Sr and is expected to have an output of 24 watts(e) and an overall efficiency of 8%. About 80% of the total electric power will be generated by the Ge–Si element. (Bölkow, Munich; and Siemens, Erlangen)

Photoelectric Conversion

The only promising isotopic fuel for photoelectric cells appears to be ^{147}Pm, whose suitability is due to its low maximum beta energy, which minimizes damage to the phosphors, its relatively long half-life (2.6 years), and its price. Tests on a mixture of ^{147}Pm hydroxide and zinc sulfide showed that the higher the specific activity of the nuclide–phosphor mixture, the stronger the drop in light output after some aging. At

present the optimum value seems to be a specific activity of about 100 curies per gram of phosphor. The light output still drops initially, but after a month it seems to remain constant. The hydroxide of ^{147}Pm was the most satisfactory compound. Irradiation of the phosphor alone by a ^{60}Co source has shown that radiation damage occurs, but this damage accounts for only a small percentage of the observed total drop in light output. Based on the results obtained to date, the filling of a photoelectric battery is under way. (Institut für Radiochemie, Technische Hochschule, Munich; and I. Physikalisches Institut der Universität, Giessen)

A photoelectric conversion cell using selenium photocells has been fabricated and was to be filled with ^{147}Pm/ZnS(Cu) phosphors in December 1967. With an overall efficiency of about 0.07%, the cell has been calculated to yield an output of 10 to 20 $\mu w/cm^2$. Another conversion system with cadmium sulfide photocells is to be built early in 1968, and it is hoped that the overall efficiency will be a factor of 2 to 5 times that with the selenium-cell device. Whether higher efficiencies will be reached depends on the availability of phosphors with the necessary high specific activity. (I. Physikalisches Institut der Universität, Giessen)

Two other possible conversion systems are considered here. In one, alpha or beta particles excite luminescence in Ga–As, which is converted in an adjacent *p-n* junction to electrical energy. The deteriorating effect of the self-absorption of the luminescence radiation can be circumvented by using a semiconductor system with a locally graded energy that exhibits the so-called "window" effect. The system $GaAs_xP_{1-x}$, with x as a mixing parameter, is particularly suitable. In the second type, there are sandwich layers of the metal oxide–metal type. In the metal layer, alpha or beta particles excite electrons that can surmount the potential barrier of the oxide. The built-in field of the potential barrier leads to a rectification effect and thus to a net unidirectional current. Suitable oxides are those of tantalum and niobium produced by anodic oxidation. The system should show very good radiation-resistance properties. (Technische Hochschule, Munich, Institut für Technische Elektronik)

Thermophotovoltaic Conversion Systems

A system is being considered in which the radiation emitted by a body at temperatures between 1200 and 1700°C can be converted to electrical energy with an efficiency of more than 10% using a photovoltaic element. The photovoltaic element is proposed to be made of a semiconductor whose energy gap is graded in the space-charge region. Such systems can be realized by using mixed-compound semiconductors with variable composition, e.g., $Ga_xIn_{1-x}As$. (Battelle Institut, Frankfurt/Main)

Pacemakers

The average lifetime of implanted cardiac pacemakers today is about 20 months. The conditions to be fulfilled by these devices are 70 to 80 pulses per minute of 2 msec length at 20 μa and 6 volts. With an isotopic battery instead of a conventional Hg/ZnO battery, pacemakers with lifetimes of about 10 years should be possible. Development work is in progress for an isotopic battery powered by 50 mg of ^{238}Pu enclosed in a cylindrical capsule with a semiconductor thermoelement conversion system. Fifty milligrams was chosen as the theoretical minimum amount for an isotopic heat source corresponding to 20 mw(t) which must provide about 120 μw at 5 volts per pulse at an overall efficiency of 1%. (Siemens, Erlangen) (MG)

Section
VI
Isotopes and Radiation Technology

Applications in Life Sciences

Tagging of Small Animals with Radioisotopes for Tracking Purposes*

By Martha Gerrard

Abstract: *A review of the literature (32 references) shows that minute radioisotope sources attached externally to or inserted subcutaneously in small animals may be used for tracking purposes.*

For many years the standard methods of marking small animals for tracking purposes have been mutilation (ear or toe clipping), tagging (banding or ringing), and coloring (dyeing or painting).[1] Unfortunately, these markings cannot be readily observed, at times when they are most needed, in burrowing and nocturnal animals. The radiotransmitters more recently used with some larger animals are usually too heavy for mice and bats, and live trapping is a time-consuming procedure. Therefore, when radioisotopes began to be used for tracing purposes in the 1950's, it was natural that they should also be applied in the field of ecology. With radioisotope tracking techniques, animals can be studied under natural conditions, without human interference, and can be followed under a layer of debris or earth or at night. Radioactive tracers are easily applied and do not affect the animal's attraction to predators. A disadvantage of this labeling technique is that the radiation detector used to track the animal actually traces the label and does not distinguish between different animals. Also, a radioactive tag on a live animal produces a finite biological effect, which must be minimized by minimizing all factors that contribute to radiation dose. However, the technique is accurate, and it permits studies that might otherwise be impossible.[2]

The literature search summarized here was made primarily to learn what radioisotopes have been used as discrete sources for tagging small mammals, but a few random papers describing discrete-source tagging in other species were included. The list of references is undoubtedly incomplete since the search was made by title only and in a limited number of journals.

Technique

When radioisotopes are used in the tracking of animals, they are usually deposited or attached to a band or ring that is fastened to the animal externally, or a fine wire or capsule containing the radioactive material is inserted just under the skin. The animal's movements are then followed with a radiation detector.

The radioisotope tracer selected should be one that can be used in amounts small enough to do negligible harm to the animal but still be readily detected by standard detection equipment. Its gamma radiation should be intense enough to be detected several feet away from the animal and through several inches of soil or litter. The beta radiation should be as low as possible since it can produce biological effects even though it does not contribute to detection. The radioisotope half-life should be short so that the activity will decay rapidly and cause no hazard if the tag is lost, and yet long enough for the radioisotope to be readily detected for the duration of the experiment.

Several radioisotopes that have been used in animal tracking are given in Table 1. The question of how much radiation can be safely used on small animals is discussed at length by Griffin.[3a] Barbour and

*Stephen V. Kaye, of the ORNL Health Physics Division, kindly reviewed the manuscript of this paper and supplemented the reference list.

Table 1 Radioisotopes Used in Animal Tracking

Radionuclide	Half-life	Radiation energies of interest, Mev	
		Beta	Gamma
^{60}Co	5.3 years	0.31 (99%)	1.33 (100%) 1.17 (100%)
^{198}Au	2.7 days	0.96 (99%)	0.41 (95%)
^{131}I	8 days	0.61 (86%) 0.33 (9%)	0.36 (82%) 0.64 (7%)
^{124}Sb	60 days	2.31 (21%) 1.60 (7%) 0.97 (9%) 0.61 (49%) 0.24 (14%)	0.60 (97%) 0.64 (7%) 1.69 (50%) 2.08 (7%)
^{182}Ta	115 days	0.5 (67%)	1.2 (13 to 34%)

Harvey[3b] consider that the radioactive source tag does not affect the animal's behavior.

Animals Studied

Among the small mammals that have been studied by the radioisotope tracking procedure are voles, moles, mice, bats, and shrews. These are all animals whose movements have to be followed at night, beneath soil or litter, or behind walls.

Voles

Godfrey,[4] in a 5-month study in 1951, used ^{60}Co to locate field vole (*Microtus agrestis*) nests, some of which were partially covered with earth or litter. She first captured a number of animals in live traps and labeled each pregnant female with 100 μc of ^{60}Co, which was in wire form and enclosed in a brass tube. A ring made from one of these tubes was fastened on the leg of each animal, just above the ankle, and the animals were released. Each day, the probe of a portable Geiger–Mueller counter was passed over the territory where the nests were thought to be, and they were actually found where the detector showed high readings.

Barbour[5a] used ^{60}Co to determine the amount of time spent in the nest by prairie voles (*Microtus ochrogaster*). He inserted 55 μc of the tracer, as a very fine wire, under the skin of the animal and located a G–M counter over the nest. Thus, whenever the animal was in the nest, activity was recorded. The home range of similarly tagged voles was determined by following them with a scintillation counter.[5b]

Iodine-131 was used by Johanningsmeier and Goodnight[6] in a 2-week study of the movements of *Microtus pennsylvanicus*. The tracer, ^{131}I dissolved in sodium sulfite solution, was placed in a double-walled capsule (0.62 in. OD) and inserted under the skin through a 12-gage hypodermic needle, using a wire plunger to push the capsule through the needle. The animal's movements were followed with a G–M counter on the end of an 8-ft-long pole. With a tag of 0.5 mc of ^{131}I, an animal could be located from a distance of 3 to 4 ft. Of course, when the animal was several inches under the ground, accurate detection was not possible.

Graham and Ambrose[7] chose ^{182}Ta for a 3-week study of *Microtus pennsylvanicus*. The tracer, as 1-cm-long by 18-gage wire, was placed under the skin of an anesthetized animal, and the animal was released in an enclosure. Its movements were followed by a NaI(Tl) crystal coupled to a photomultiplier tube.

Moles

The activity and rest periods of moles (*Talpa europaea*) were studied by Godfrey,[8] using a method similar to that used for voles.[4] In this case, 80 μc of ^{60}Co was used in Monel rings. Since the shape of the mole leg makes leg rings impractical, the rings were located at the base of the tail. A G–M counter was used to follow the animals in this 2-month study. The animals moved between 20 and 830 yd and could be detected when 1 ft beneath the ground surface.

Mice

Gold-198, in the form of subcutaneous wires, was used by Kaye[9] in studying the movements of Eastern harvest mice (*Reithrodontomys humulis humulis*). Each animal was tagged with 0.7 to 4.5 mc of the radioisotope, which was inserted under the skin as a 10-mm-long 20-gage wire. A G–M counter was used to track the animals. With a 0.7-mc tag an animal was detectable for 1 week, initially at a distance of 9 ft but decreasing to 1.5 ft by the seventh day. With larger amounts of radioisotope, the initial distance at which the animal could be detected was up to 20 ft.

Orr[10] used ^{182}Ta wire in mice to determine their behavior under field conditions prior to starting a study of the effects of irradiation on home-range behavior. Inglis et al.[11] used 0.5 mc of ^{182}Ta wire (3 mm long, 16 gage) subcutaneously to determine the time spent in its den by a male mouse (*Baiomys*

taylori). They used a portable scintillation detector to locate the animal in its burrow and then placed a G–M probe on the ground over the spot. With this arrangement the experimenter did not have to be present at all times.

Bats

Gifford and Griffin[12] used ^{131}I in studies on bats (*Myotis lucifugus* and *Eptesicus fuscus*) and were able to locate the animals through the walls of buildings. They used bands prepared by dropping ^{131}I solution, with an activity of 5 μc, on one end of the band, evaporating the solution, and covering the radioactive deposit with a thin layer of glue. The end of the band was then folded over the deposit and crimped. The ^{131}I half-life was long enough for this experiment since the chief interest was in the number of animals that returned home during the first few days after being released at a distance.

Punt and Van Nieuwenhoven[13] used ^{124}Sb for an experiment lasting several months on 11 species of bats that hibernated in limestone caves. Each bat was labeled with 250 μc of radioactive tracer in the form of a 1-mm-diameter 2-mm-long wire weighing 10 mg. The wire was placed in a small tube at the end of an aluminum band (4 mm wide, 120 mg weight), the tube being formed by rolling back the end of the band and closed by crimping. The bats could be located with a G–M counter even when hiding deep in a crooked fissure; this was not possible by visual methods. The amount of activity used was considered small enough that no harm would be done to the animal if it should not be recovered, and no activity would be carried over to the next season to interfere with future experiments.

Davis, Barbour, and Hassell,[14] in a manner similar to the earlier work of Harvey and Barbour,[5b] inserted ^{60}Co wires under the forearm skin of big brown bats (*Eptesicus fuscus*) for studying colonial behavior.

Shrews

Linn and Shillito[15] modified the Godfrey[4] ring to reduce the weight and thus make it suitable for use on shrews. The new ring had an activity of 50 μc. They also mention a silver ring, which was made radioactive by reactor irradiation to form ^{110m}Ag (270 days).

Other Animal Species

Naulleau and Courtois[16a] readily detected snakes (*Vipera aspis*), both on the ground and to a depth of about 1.5 ft, which they had labeled with 90 μc of ^{60}Co. By means of a hypodermic needle, they injected a 0.5-mm-diameter 1-cm-long wire subcutaneously in the dorsal region of the tail. They followed the animals' movements with a scintillation counter. That the tagging was not harmful to the snake was indicated by the difficulty experienced in locating the wire in some of the recovered animals; in others there was a small scar at the injection site. With 14 μc of ^{60}Co, detection was slow, and with 0.5 and 1 mc, the animals were definitely harmed. In later work,[16b] the ^{60}Co wire was enclosed in plastic, and 45 μc was used. These snakes were in perfect health a year later.

Karlstrom[17] tagged frogs with ^{60}Co inserted subcutaneously. A solution of the radioisotope was placed in a hollow drilled in a lead shot and evaporated. The shot was crimped and pressed into a tablet, filed smooth, and sprayed with plastic. Both a G–M and a scintillation counter were used for detection. Various conditions were tested, and a 20-μc tag was found to be locatable 4 ft away when it was on the surface of the ground, for example. One tag was recovered that was buried 15 in. below the surface. Breckenridge and Tester[18] inserted a ^{182}Ta wire (1 mm diameter, 5 mm long, 100 μc activity) under the skin of the anterior dorsal region of frogs and followed them with a scintillation counter. Animals were detected 20 ft away on the surface, and one frog with a 4-month-old tag was found 22 in. deep in the soil.

At the University of Minnesota, William Nelson (personal communication quoted in Ref. 2c) used 100 μc of ^{182}Ta subcutaneously in skinks (*Eumeces septentrionalis septentrionalis*). The wires were 0.020 mm diameter by 5.0 mm long. O'Brien, Smith, and Meyer,[19] in studying the movements and behavior of lizards (*Sceloporus undulatus hyacinthinus*), used 1 mc of ^{198}Au as an 18-gage wire (5 mm long) enclosed in polyethylene tubing. They attached the tag externally on the ventral surface of the animal by tying it in place with 10 cm of No. 8 silk thread. They could detect the animal with a G–M survey meter (10-ft probe lead) initially at 12 ft but after 7 days, at only about 5 ft.

Griffin[3] used ^{65}Zn for tagging birds. The radioisotope was placed in an aluminum capsule attached to a conventional bird band. A battery-powered G–M counter buried at the side of a bird nest recorded the time of return of a tagged bird to its nest with an accuracy of ±5 min. The equipment operated, unattended, for 8 days under summer Arctic field conditions. However, Tester[2c] mentions a personal communication that indicates dissatisfaction with the method. Blue grouse (*Dundragapus obscurus*) tagged

with 500 μc of ^{86}Rb on standard patagial wing bands could not be followed to their nests. The range of detection with a G–M counter was 5 to 10 ft. He also refers to a somewhat different technique, in which a ^{90}Sr source was placed on one side of the nest (mallards, *Anas platyrhynchos platyrhynchos*) and a radiation detector on the other. The radiation beam was interrupted when the bird was in the nest. In still another method, ^{45}Ca (as calcium phosphate) was inserted as pellets in the breasts of ring-necked pheasants (*Phasianus colchicus*), and the progeny of the birds were traced by the radioisotope deposited in their bones. Neither of these last two techniques, of course, is really an inserted-source technique.

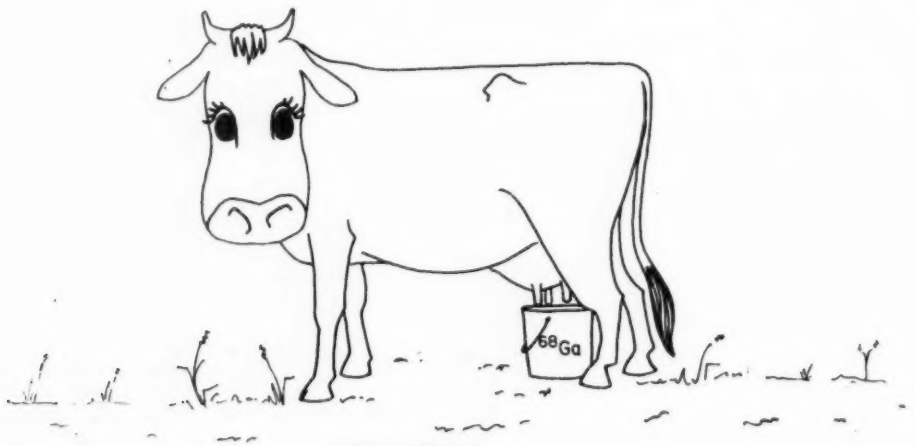
Radioisotope sources have been inserted in the abdominal cavities of fish. Hiyama[20] refers to the use of coated ^{60}Co pins (0.1 to 1 mc). He also considers the public health problem of tagged fish being used for food, both those tagged with a source and those labeled by diffusion of the isotope throughout the body, e.g., ^{65}Zn. Wilimovsky[21] used Alnico V magnetic rods (0.5 in. long, 0.125 in. diameter), which were made radioactive (10 μc) by reactor irradiation to produce ^{60}Co and coated with polyethylene. His studies were with Alaska herring (*Clupea harengus pallasi*), and the tagged fish were recovered from the commercial catch when delivered to the processing plant. Seymour,[22] however, after reviewing various methods of tagging fish, concluded that radioisotope tagging is not practical in this field.

Although insects are usually tagged with a radioisotope that is diffused throughout the organism (e.g., by allowing the larva to develop on a labeled nutrient medium), a discrete source may be used with a large insect. Tomes and Brian[23] labeled beetles with 5 μg of radium sulfate, which was deposited between aluminum-foil disks. The disks were inserted, with resin adhesive, beneath the elytra, and the insects were followed with a G–M counter. Arnason, Fuller, and Spinks[24] placed 2 mg of radioactive cobalt (^{60}Co) in the caudal notch of the larvae of prairie grain wireworms [*Ctenicera aeripennis destructor* (Brown)], securing it with plastic. The insects were followed with G–M counters.

References

1. *a.* R. D. Taber, Uses of Marking Animals in Ecological Studies: Marking of Mammals; Standard Methods and New Developments, *Ecology*, **37**: 681-685 (1956).
   b. G. C. Sanderson, The Study of Mammal Movements: A Review, *J. Wildlife Manage.*, **30**: 215-235 (1966).
2. *a.* R. C. Pendleton, Uses of Marking Animals in Ecological Studies: Labeling Animals with Radioisotopes, *Ecology*, **37**: 686-689 (1956).
   b. G. D. Godfrey, Use of Radioactive Isotopes in Small-Mammal Ecology, *Nature (London)*, **174**: 951-952 (1954).
   c. J. R. Tester, Techniques for Studying Movements of Vertebrates in the Field, in *Radioecology*, Proceedings of the First National Symposium on Radioecology, Colorado State University, 1961, pp. 445-450, Reinhold Publishing Corp., New York, 1963.
3. *a.* D. R. Griffin, Radioactive Tagging of Animals Under Natural Conditions, *Ecology*, **33**: 329-335 (1952).
   b. R. W. Barbour and M. J. Harvey, The Effect of Radioactive Tagging on the Activity of Mammals, *Amer. Midland Natur.*, **79**: 519-522 (1968).
4. *a.* G. D. Godfrey, Tracing Field Voles (*Microtus agrestis*) with a Geiger–Mueller Counter, *Ecology*, **35**: 5-10 (1954).
   b. G. D. Godfrey, A Technique for Finding *Microtus* Nests, *J. Mammalogy*, **34**: 503-505 (1953).
5. *a.* R. W. Barbour, *Microtus:* A Simple Method of Recording Time Spent in the Nest, *Science*, **141**: 41 (1963).
   b. M. J. Harvey and R. W. Barbour, Home Range of *Microtus ochrogaster* as Determined by a Modified Minimum Area Method, *J. Mammalogy*, **46**: 398-402 (1965).
6. A. G. Johanningsmeier and C. J. Goodnight, Use of ^{131}I To Measure Movements of Small Animals, *Science*, **138**: 147-148 (1962).
7. W. J. Graham and H. W. Ambrose III, A Technique for Continuously Locating Small Mammals in Field Enclosures, *J. Mammalogy*, **48**: 639-642 (1967) (presented at Meeting of American Society of Mammalogists, Long Beach, Calif., 1966; CONF-660652-1).
8. G. D. Godfrey, A Field Study of the Activity of the Mole (*Talpa europaea*), *Ecology*, **36**: 678-685 (1955).
9. *a.* S. V. Kaye, Gold-198 Wires Used To Study Movements of Small Mammals, *Science*, **131**: 824 (1960).
   b. S. V. Kaye, Movements of Harvest Mice Tagged with ^{198}Au, *J. Mammalogy*, **42**: 323-337 (1961).
10. H. D. Orr, Effects of Irradiation on the Behavior of Small Mammals Under Field Conditions, USAEC Report COO-1406-2, St. Olaf College, September 1967 (*Nucl. Sci. Abstr.*, **22**: 14979).
11. J. M. Inglis, L. J. Post, C. W. Lahser, and D. V. Gibson, A Device for Automatically Detecting the Presence of Small Animals Carrying Radioactive Tags, *Ecology*, **49**: 361-363 (1968).
12. C. E. Gifford and D. R. Griffin, Notes on Homing and Migratory Behavior of Bats, *Ecology*, **41**: 378-381 (1960).
13. A. Punt and P. J. Van Nieuwenhoven, The Use of Radioactive Bands in Tracing Hibernating Bats, *Experientia*, **13**: 51-54 (1957).
14. W. H. Davis, R. W. Barbour, and M. D. Hassell, Colonial Behavior of *Eptesicus fuscus*, *J. Mammalogy*, **49**: 44-50 (1968).
15. I. Linn and J. Shillito, Rings for Marking Very Small Mammals, *Proc. Zool. Soc. (London)*, **134**: 489-495 (1960).

16. *a.* G. Naulleau and G. Courtois, Utilization of ^{60}Co for the Marking of Snakes (in French), *Compt. Rend.*, **260:** 6219-6222 (1965).
b. G. Naulleau, Technique for Labeling Snakes with Radioisotopes (in French), *Compt. Rend., Ser. D*, **265:** 1977-1979 (1967).
17. E. L. Karlstrom, The Use of ^{60}Co as a Tag for Recovering Amphibians in the Field, *Ecology*, **38:** 187-195 (1957).
18. W. J. Breckenridge and J. R. Tester, Growth, Local Movements and Hibernation of the Manitoba Toad, *Bufo hemiophrys*, *Ecology*, **42:** 637-646 (1961).
19. G. P. O'Brien, H. K. Smith, and J. M. Meyer, An Activity Study of a Radioisotope-Tagged Lizard, *Sceloporus undulatus hyacinthinus* (Sauria: Iguanidae), *Southwestern Natur.*, **10**(3): 179-187 (1965).
20. Y. Hiyama, Radioisotope Marking Methods of Fish and the Public Health Consideration (in Japanese, English abstract), *Radioisotopes (Tokyo)*, **11:** 389-393 (1962).
21. N. J. Wilimovsky, An Application of Radioisotopes in Fish Tagging, *Proc. Alaskan Sci. Conf.*, **11:** 93-99 (1961) (quoted in Ref. 2*c*).
22. A. H. Seymour, The Use of Radioisotopes as a Tag for Fish, *Proc. Gulf and Caribbean Fish. Inst.*, 10th Annual Session, pp. 118-124 (1957) (quoted in Ref. 2*c*).
23. G. A. R. Tomes and M. V. Brian, An Electronic Method of Tracing the Movements of Beetles in the Field, *Nature (London)*, **158:** 551 (1946).
24. A. P. Arnason, R. A. Fuller, and J. W. T. Spinks, An Electronic Method of Tracing the Movements of Soil-Inhabiting Insects, *Science*, **111:** 5-6 (1950).



Isotope "Cow"

Section
VII
Isotopes and Radiation Technology

Isotopes Around the World

Recent Isotopic Source Developments in the USSR

By Martha Gerrard

Among the interesting radiation-source developments in the USSR are (1) a facility that uses spent fuel elements for vulcanizing tires; (2) a facility with a cone-shaped source for modifying the properties of similarly shaped plastic parts, for example; and (3) a new isotopic generator in the "Beta" series. In addition, a state-of-the-art review of radiation-chemical processing provides a handy bibliography of both USSR and non-USSR work in this field prior to its date of publication (1964). These developments are summarized below. Several USSR source developments described in earlier issues of *Isotopes and Radiation Technology* are the first USSR industrial radiochemical plant,[1] which uses ^{60}Co in the manufacture of sulfonate detergents; a commercially produced mobile irradiator,[2] which uses ^{137}Cs to irradiate seeds prior to sowing; and the gamma irradiator of the L. Ya. Karpov Physical Chemistry Institute.[3]

Spent-Fuel-Element Source for Vulcanizing Tires

A pilot plant for vulcanizing tires with the radiation from spent fuel elements was operated[4] to obtain data on the feasibility of industrial application of the process. The unit, with 18 spent fuel elements as the radiation source, was installed in a large-scale facility[5] that had a capacity for handling a source made from 100 such elements. When not in use, the source was stored in a water-filled pool (Fig. 1).

In the vulcanization pilot plant unit, six small automobile tires, separated by metal gaskets to prevent warping, were placed in a cylindrical tank located at

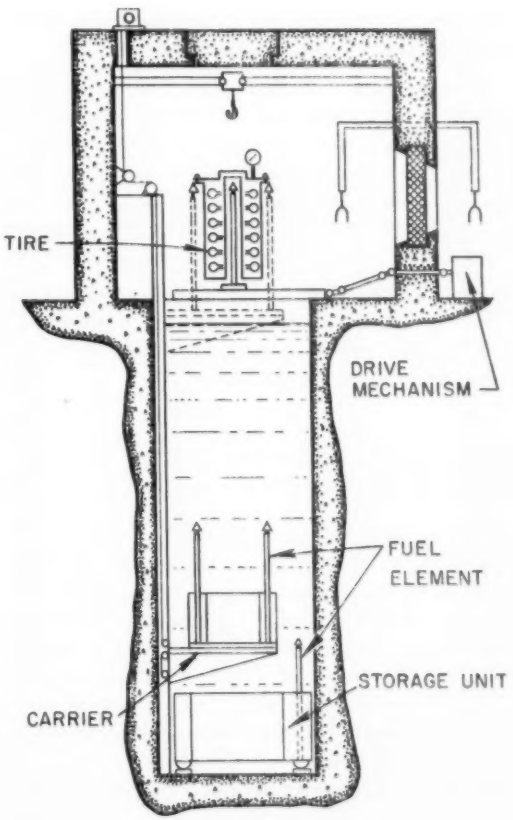


Fig. 1 Experimental radiation-chemical vulcanization facility using spent fuel elements.

the surface of the water. In preparation for an irradiation, the tank was closed and the atmosphere replaced with argon at slightly above atmospheric pressure. Then the source, in which the 18 fuel elements were arranged circumferentially on a 90-cm-diameter carrier, was raised from the bottom of the pool and positioned so that the tank containing the tires was surrounded by the fuel elements. The total radiation dose to the tires is not stated in the original article, but the average dose rate of about 50 r/sec is said to permit processing about 1300 tires per year in this facility.

The gamma-radiation field from the spent fuel elements varied (Fig. 2); hence, to ensure that doses to the tire treads varied no more than ±10%, the first and third tires and the fourth and sixth tires were shifted in position at the midpoint of the irradiation time. Thus about 80% of the height of the irradiator could be used. To provide a homogeneous field in the horizontal plane, the tire assembly was rotated about the vertical axis at a rate of 3 rpm during the irradiation.

The individual fuel elements had been stored 160 days after reactor discharge, and their activities varied from 2600 to 5100 curies because of their having been in different locations in the reactor. The initial total activity of the 18-element assembly was 76 thousand curies. Because the activity of these elements decreases rather rapidly, dose rates were measured every 2 weeks.

The efficiency of radiation utilization in this facility was about 3%, but it could be increased to 50% or more in an industrial setup by proper location of source and objects to be irradiated.

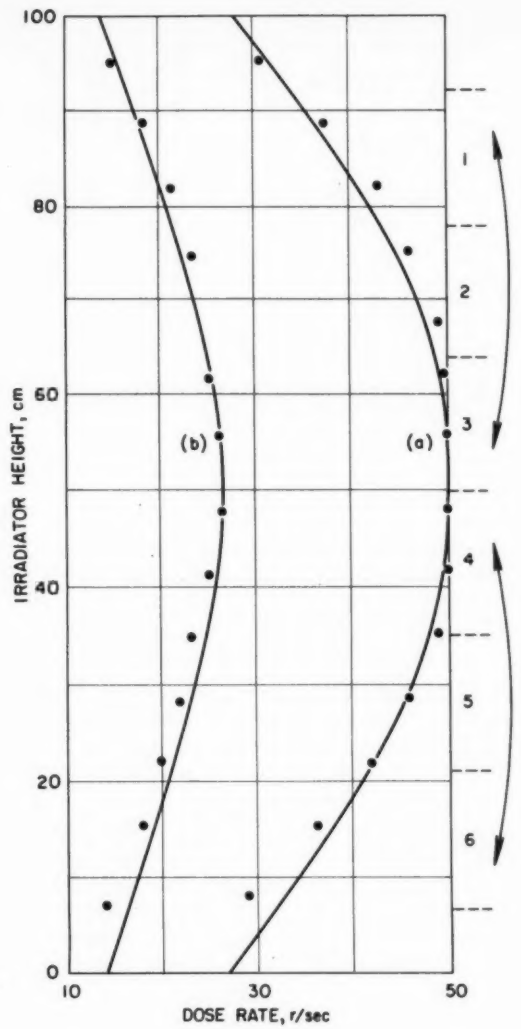


Fig. 2 Experimental (points) and calculated (lines) rate of irradiation of tire treads for different heights of irradiator filled with automobile tires. Fuel elements held (*a*) 160 days and (*b*) 260 days after withdrawal from reactor. Numbers on right indicate shifting of tires to equalize dose.

Gamma-Irradiation Facility with Cone-Shaped Source

The gamma-irradiation facility[6] at the L. V. Pisarzhevski Physical Chemistry Institute of the USSR Academy of Sciences was designed for irradiation, at a dose rate of 470 rads/sec, of conical objects made of plastic or metal. The source (Fig. 3), which contains 400 to 1200 curies of ^{60}Co, is fabricated from 12-mm-ID stainless-steel tubes in which the radioactive material is placed. The upper part of the irradiator is cylindrical and consists of 16 tubes, 95 mm long, which are equally spaced around a 152-mm-ID circle. The lower part is cone-shaped and is made of 4 tubes that are 250 mm long and 12 tubes that are 170 mm long. These tubes are suspended with some freedom of movement in the vertical plane. The angle of the cone, and hence the dose rate in the conical portion, can be varied by moving the restraining ring up or down. The tubes for the cylindrical and conical sections of the source are loaded separately and then connected.

The irradiator is bolted to the bottom of a deep pool of water (Fig. 4), which acts as a shield. Objects to be irradiated are placed in a metal container attached to a long rod and lowered into the irradiator.

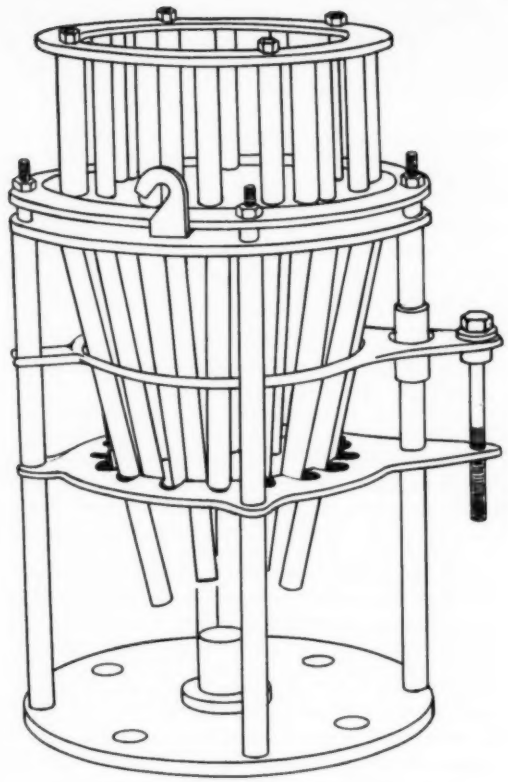


Fig. 3 Russian cone-shaped ^{60}Co irradiator.

"Beta" Series of Isotopic Power Sources

A series of isotopic power sources, designated the "Beta" series, has been developed[7] in the USSR for use at unattended weather stations. In these generators the heat released in radioactive decay is converted to electricity for powering the equipment.

The first generator[8] in this series, the Beta-1, was constructed and put into operation at a weather station in 1963. It contained 17.5 kilocuries of 284-day ^{144}Ce + ^{144}Pr and had a nominal power output of 5 watts. It operated for 8800 hr and produced 25 kw-hr of electricity during this time.

A year later, a Beta-2 model was constructed.[9] This generator used ^{90}Sr and had a nominal power output of 5 to 7 watts. The long half-life (28 years) of ^{90}Sr allowed it to operate 30 thousand hr. A weather station using a generator of this type could probably operate 5 to 10 years without maintenance on the power source. The Beta-C generator, constructed in 1967, is similar to Beta-2 but has a nominal power output of 10 watts and should allow 10 years' unattended operation. The Beta-3, in turn, is similar to Beta-C but is designed for operation under conditions in the Far North or the Antarctic, i.e., at −70°C.

A very recent development[10] in the Beta series is the MIG-67, an improved model of Beta-C, which uses ^{238}Pu. Designed for laboratory use, it is said to weigh only 11 lb and to have a nominal power output of 1 watt.

State of the Art of Radiation Processing

A Russian book[11] on radiation-chemical processing, printed in 1964, indicates the considerable work being done in the USSR on the use of nuclear

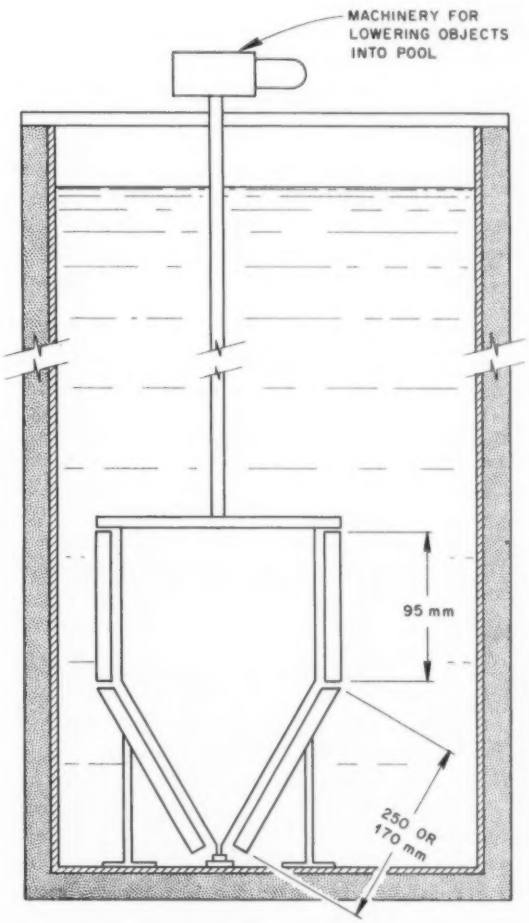


Fig. 4 Cone-shaped ^{60}Co source installed on the bottom of a water-filled pool.

radiation sources. Persons who do not read Russian will find the bibliography (approximately 600 references) useful as a guide to both Russian and non-Russian work. Those who read Russian will find it useful because of the large amount of basic information assembled in one place.

Basic information discussed includes the properties of potentially useful radiation sources—natural and artificial radioisotopes, accelerators, and reactors. Then the gamma sources ^{60}Co and ^{137}Cs are compared, with reference to both stationary and mobile equipment, and beta sources are discussed briefly. Designs of a number of large- and small-scale gamma-irradiation installations and of reactor-loop arrangements are shown. Finally, methods for calculating the basic parameters for radiation-chemical apparatus and safety regulations to be observed in radiation-chemical processing plants are discussed in detail. Some of the Russian code designations for irradiation equipment are identified, and several USSR and non-USSR installations are sketched.

References

1. I. F. Sprygaev and S. V. Mamikonyan, First Industrial Radiochemical Plant in the USSR (in Russian), *At. Energ. (USSR)*, **23**: 469-470 (1967) [*Isotop. Radiat. Technol.*, 6(1): 114 (Fall 1968)].
2. B. Zhukov, Commercial Gamma-Ray Sources for Pre-Sowing Irradiation of Seeds (in Russian). *At. Energ. (USSR)*, **23**: 477-478 (1967) [*Isotop. Radiat. Technol.*, 6(1): 114-115 (Fall 1968)].
3. ^{60}Co Irradiation Facility of the L. Ya. Karpov Physical Chemistry Research Institute (in Russian), *At. Energ. (USSR)*, **18**: 546-548 (1965) [*Isotop. Radiat. Technol.*, 5(1): 59-60 (Fall 1967)].
4. V. A. Borisov, V. E. Drozdov, G. N. Lisov, Yu. S. Ryabukhin, L. M. Suroegin, V. E. Lesnichii, I. M. Zakharova, A. G. Vasil'ev, and L. M. Dunaev, Pilot-Plant Irradiation Facility Using Spent Reactor Fuel Elements for Thermo-Radiation Vulcanization of Tires (in Russian), *At. Energ. (USSR)*, **23**: 141-142 (1967).
5. V. L. Karpov, A. Kh. Breger, M. E. Eroshov, V. E. Drozdov, G. N. Lisov, S. G. Stoenko, D. M. Torgovitskii, B. I. Vainshtein, and N. P. Syrkus, Large-scale Radiation-Chemistry Facility for Irradiation with Spent Fuel Elements (in Russian), *At. Energ. (USSR)*, **15**: 302-308 (1963).
6. G. N. P'yankov and M. A. Brashkin, Gamma-Irradiation Facility with Complex Source (in Russian), *At. Energ. (USSR)*, **24**: 192-193 (1966).
7. G. M. Fradkin, V. M. Kodyukov, A. I. Ragozinskii, E. A. Kazakov, N. P. Korotkov, and D. Ya. Surazhskii, Isotopic Sources of Electricity for Unattended Meteorology Stations (in Russian), *At. Energ. (USSR)*, **24**: 203-204 (1968).
8. A. N. Voronin, I. G. Gverdtsiteli, R. Z. Grinberg, Yu. D. Gubanov, V. A. Zharkov, V. F. Zhukov, V. K. Kovyrzin, V. M. Kodyukov, N. P. Korotkov, R. Ya. Kucherov, S. P. Lalykin, A. I. Ragozinskii, G. M. Fradkin, B. L. Shinderov, and S. Ya. Erman, Thermoelectric Generators with Isotopic Heat Sources (in Russian), in *Proceedings of the Third International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1964*, Vol. 15, pp. 176-184, United Nations, New York, 1965 [extracted in *Isotop. Radiat. Technol.*, 2(3): 282-284 (Spring 1965)].
9. G. M. Fradkin, V. M. Kodyukov, and A. I. Ragozinskii, Isotopic Source of Electrical Energy, Beta-2 (in Russian), *At. Energ. (USSR)*, **18**: 545-546 (1965).
10. *Nucleonics Week*, June 6, 1968, p. 8.
11. A. Kh. Breger, B. I. Vainshtein, N. P. Syrkus, V. A. Gol'din, and L. V. Chepel', *Principles of Construction of Radiochemical Facilities* (in Russian), Atomizdat, Moscow, 1964 (498 pp.).

Practical Applications of Radioisotopes in the USSR

L. Kostikov recently reviewed[1] the economic savings resulting from the use of radioisotopes in the USSR. His work was briefly summarized elsewhere.[2] Kostikov is the former head of the Industrial Research Laboratory for Heat Transfer Problems in Power Generation at the Bauman Higher Technical Institute in Moscow; he is currently a Project Officer with UNESCO in Paris.

Kostikov discusses the uses of radioisotopes in the USSR under four headings: isotopes in process monitoring and measurement; tagged atoms; isotopes in agriculture, biology, and medicine; and isotopes in radiation chemistry.

Isotopes in Process Monitoring and Measurement

All measuring or monitoring instruments based on radioisotopes consist of a radiation source, a detector, an amplifier, and an impulse counter. In coating-thickness measurements, beta particles from a radioisotope are reflected from the inner surface of the coating to the detector; the material of the coating must be at least two or three atomic numbers different from the material covered. Tin plated on sheet iron can be measured at a thickness of only 5 μ to within an

accuracy of 0.1 μ. The USSR Academy of Sciences' Institute of Economics found that beta gages in industry result in average savings of 28,500 rubles* per year.

Radioisotope weight-measuring devices operate on the principle that the ability of a material to absorb gamma rays depends on its density and thickness. The weight of railroad cars† is measured by an installation consisting of a ^{60}Co source under the railway track and a scintillation counter above; the moving train diminishes the intensity of the radiation reaching the counter according to its weight.

A device used at the Krasnogorsk paper mills helps produce paper weighing 50 g/m^2 with a tolerance of ±2%. The USSR State Standard is ±5%. Each paper-making machine at Krasnogorsk can thus produce an additional 2100 kg of paper every 24 hr. In general, radioisotope devices measuring sheet material thickness and coatings save tens of millions of rubles every year.

Level-measuring devices are widely used in the metallurgical industry. Accurate data on the level to which the blast furnace has been charged with coke make it possible to maintain optimum operating conditions. At the Dzershinsky and Novotulsky steelworks, the use of multipositional level-measuring instruments has increased production 2% and decreased costs 1% for each ton of pig iron. Applied to all Soviet blast furnaces, radioisotope gages would save over 20 million rubles each year.

Over 2000 stationary or portable gamma defectoscopes are now in use in the USSR. Throughout Soviet industry, these instruments save over 30 million rubles each year. Portable devices using ^{170}Tm or ^{155}Eu detect flaws in steel from 1 to 15 mm thick and in aluminum from 5 to 50 mm thick. In general, a gamma photograph is cheaper than an X-ray picture by about 20 kopeks. Recently, visual inspection with scintillation screens as detectors has come into use; continuous automated inspection of products is potentially feasible with this method.

Tagged Atoms

Radioactive tracers are widely used in Soviet industry. Because this direct method of checking production techniques gives good quantitative results, new ways of obtaining high-purity Zn, Sn, Pb, Ni, Te, Tl, and other metals have been developed. At the Donetsk steelworks, output has been increased by 750,000 rubles' worth per year. Additional savings of about 90,000 rubles in overhead and of 180,000 rubles as a result of using slag in place of iron ore are also benefits of radioactive-tracer research. Interestingly, every ruble spent on process research and new production techniques saves 10 rubles.

In the USSR oil industry, radioisotopes are used to locate breaks in drilling pipes, to locate places where water enters wells, to check the condition of oil wells, to check the quality of major repairs, to determine the zone of liquid circulation around the pipe, and to determine the height and thickness of cement plugs. The Institute of Economics of the USSR Academy of Sciences calculates that radioisotope use in the oil industry saves over 30 million rubles a year.

Tagged atoms are also important in studying wear and corrosion in machines. The cutting laboratory of the All-Union Tool Research Institute has established that 30,000 rubles' worth of workmetal and high-speed tool steel is used to determine the relationship between the durability of a cutting tool and its cutting speed by ordinary methods. With radioisotopes, nearly the entire sum is saved. In addition, the radioisotope method requires as little as 2% of the time required by ordinary methods.

Radioisotopes in Agriculture, Biology, and Medicine

Radioisotope-labeled organic compounds and mineral fertilizers are used in agriculture to work out the most efficient uses of all kinds of plant nutrients. Uzbek scientists used an isotope method to check the effectiveness of a chemical spray treatment of crops that combined an insecticide to control the red spider mite on cotton plants with a super-phosphate top-dressing for the soil. According to the Uzbek Ministry of Agriculture, only 20 to 25% as much insecticide has to be used per hectare‡; thus, an area four or five times as large can be treated, with a savings of 400,000 rubles on chemicals alone. Tagged atoms have also been used to determine insecticide residue on crops and to determine the rate at which molecules of insecticides decompose within living vegetable and animal organisms.

The irradiation of seeds before sowing increased crop yields of cabbage by 5 to 10%, cucumbers 15 to

*At the official rate, 1 ruble (100 kopeks) = $1.11.

†See page 225 of this issue.

‡2.47 acres.

30%, tomatoes 10 to 15%, carrots 25 to 30%, and radishes 6 to 11%. Although these are preliminary results, the Institute of Economics of the USSR Academy of Sciences estimates that an extra 100,000 to 200,000 metric tons of vegetables could be produced on the same acreage now used. This would save 15 to 30 million rubles a year by lowering production costs.

In another experiment, after exposure to a very small dose of gamma radiation, hen eggs showed a 3.5% increase in the number that hatch; moreover, egg-laying rates of hens hatched from these eggs increased by 12 to 17%. Applied nationwide, this technique could save millions of rubles annually.

Kostikov barely mentions radioisotope uses in medicine and biology. No savings are mentioned. He describes the use of ^{131}I for thyroid treatment and diagnosis and the injection of radioactive material in liquid form to treat malignant growths.

Radiation Chemistry

Irradiated polyethylene is now a fairly well-known product. Kostikov cites ^{60}Co and ^{137}Cs as the two most commonly used radioisotopes for radiation processing. Units of hundreds of thousands of curies are now in use in the USSR, and there are plans to build units with up to 10 million curies activity. The first technical and economic trials indicate that valuable chemical products can be obtained more cheaply using large radiation sources than by any other method.

(FEM)

References

1. L. Kostikov, The Manipulation and Use of the Atomic Nucleus. II. Practical Applications of Radioisotopes, *Impact Sci. Soc.*, **18**(1): 43-49 (1968).
2. Soviet Isotopes, *Nucl. News*, **11**(6): 25-26 (June 1968).

INDONESIAN RADIOISOTOPE APPLICATION NEWS BULLETIN

*The Indonesian Radioisotope Application News Bulletin** presents brief summaries of information on uses of radioisotopes. Issue No. III (March 1968), for example, describes various gages—neutron, gamma, and X-ray fluorescence—and their use in determining moisture, thickness, levels, and other important industrial variables. Industrial savings accruing from the use of such gages are stressed. The bulletin should be effective in calling radioisotopes to the attention of industrial firms, especially in Indonesia. (MG)

*Pusat Reactor Atom Bandung, Bandung, Indonesia.

AUTORADIOGRAPHIC TECHNIQUES

Two recent books on autoradiographic techniques will be of particular interest to biologists:

W. D. Gude, *Autoradiographic Techniques*, Prentice-Hall, Inc., Englewood Cliffs, N. J., 1968 (113 pp.) (paperback, $3.95).

A. W. Rogers, *Techniques of Autoradiography*, Elsevier Publishing Company, New York, 1967 (335 pp.) (hardback, $20.00).

The book by Dr. Gude, Oak Ridge National Laboratory, is a practical one, designed primarily for persons wanting to learn to use autoradiographic techniques. The most common methods are described in detail, and information is given on how to choose radioisotopes and the amounts to be used. A number of experiments to demonstrate the technique are described, and the basic principles of autoradiography are explained. Persons wanting more detail may go to the original articles listed in the bibliography. The lists of suppliers of radioisotope kits for educational purposes and of manufacturers of products used in autoradiography, given in the appendix, will be extremely useful to the research worker or graduate student who wants to start using the technique.

The book by Dr. Rogers, Oxford (England), is a more extensive treatment of the subject and might well be considered a textbook. Each of the 18 chapters is well documented and presents a wealth of detail on such phases of the subject as autoradiographic resolution and interfering background, relative and absolute measurements of radioactivity, and details of techniques.

Both books are clearly written and have good illustrations. Many readers will want both. Other readers will select one or the other depending on whether their interest is mainly practical or mainly theoretical.

(MG)

PREPARATION OF A ^{55}Fe SOURCE BY ELECTROPLATING

By H. H. Cutshall, J. Beaver, and F. N. Case

At Oak Ridge National Laboratory, a ^{55}Fe source suitable for use in powder and single-crystal diffraction studies has been made by electroplating the radioactive metal onto a platinum-foil cathode. The radioactive film is smooth, and self-absorption of the X radiation is low (17%).

The ^{55}Fe was made by proton bombardment of ^{55}Mn in the ORNL 86-in. cyclotron. The iron, together with the manganese target layer and a small amount of the aluminum backing, was dissolved in concentrated nitric acid and the solution evaporated. The residue, largely MnO_2, was dissolved in 9*M* HCl, and the ^{55}Fe was recovered from the acid solution by extraction with dichloroethyl ether and stripping into water. Three extraction-stripping cycles were required to completely separate the iron from manganese and aluminum; the ^{54}Mn that was also formed during the irradiation was used as a tracer to determine when the separation from manganese was complete.

The ^{55}Fe thus obtained was dissolved in 2 ml of saturated ammonium oxalate, 0.15*M* in H_2SO_4. This solution was electrolyzed in a Plexiglas cell (Fig. 1), and the iron was deposited on a 2- by 10-mm platinum-foil cathode. With a 50-ma current, 95% of the iron was deposited in 90 min. (MG)

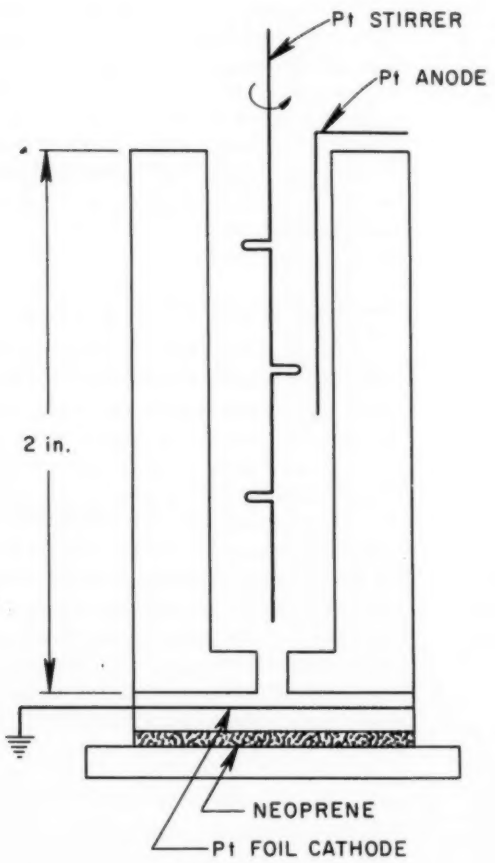


Fig. 1 Plexiglas cell for electrolytic deposition of ^{55}Fe in source preparation.

Section

VIII

Isotopes and Radiation Technology

Miscellaneous

Availability of Isotopes and Services

Nuclear Medicine Institute in Ohio

The Nuclear Medicine Institute of Mayfield Heights, Ohio (6760 Mayfield Road, P. O. Box 24062, ZIP Code 44124) is a nonprofit foundation established in 1966 to introduce physicians to nuclear medicine. It is currently offering to physicians a 54-hr (1 week) course once a month, 10 months of the year. The course includes study of radiation physics, health physics, basic instrumentation, and techniques as well as interpretation of all well-established laboratory techniques, function tests, and imaging procedures.

A 52-week course is also offered to registered technicians or nurses or to persons with degrees in biology, chemistry, or physics (with exceptions at discretion of the director). This course includes didactic and clinical training at the Nuclear Medicine Institute and clinical training at an associated hospital.

UT Adds Nuclear Medicine at Memphis

The University of Tennessee Medical Unit in Memphis plans to develop an area-wide program in nuclear medicine. The program head, Dr. Ben I. Friedman, will have the assignment of developing a teaching and training center that will be regional in nature as well as a school for technologists in nuclear medicine.

Australian School of Nuclear Technology

The Australian School of Nuclear Technology, which has been in operation since 1965, is now conducting two courses for graduate students and two for nongraduate students each year. Applicants for the graduate level courses must have a degree from an approved institution in physical science, engineering, or a related field and a fluent command of oral and written English. Selection of participants is based on the qualifications of the individual applicant and the relative requirements of his sponsor for people educated in nuclear technology.

Course work includes study of nuclear theory, interaction of radiations with matter, radiation detection, and radiological safety and laboratory work using instruments, special equipment, and techniques. The work is designed to give scientific, engineering, and technical personnel, within a short time, sufficient proficiency in radioisotope techniques and applications to use radioisotopes safely and efficiently in their individual fields.

New Bibliography Service Available

A current literature awareness service is now available from the Western New York Nuclear Research Center. Three bibliographies are being published each month. The categories and their definitions are: (1) Tracer Applications, which includes nuclear methods of analysis such as activation analysis, radiation detection, hot-atom chemistry, radioisotope production and decay schemes, tracer studies, and Mössbauer studies; (2) Radioisotope Applications, which includes instruments and applications using sealed sources, dosimetry, backscatter techniques, radiography, and isotopic power applications; and (3) Radiation Processing, which includes effects of radiation on materials, use of radiation to induce chemical reactions, and methods used to investigate radiation effects. All three categories exclude medical applications, health physics, power reactors, and mass and emission spectrometry. A typical compilation will include about 500 references in the three bibliographies. Articles and reports are listed alphabetically by author and by key word, and an order list is included.

The program is sponsored by the U. S. Department of Commerce and The New York State Technical

Services Program. Format changes and charge reductions are planned. For further information on the service, write R. T. Smokowski, Nuclear Science and Technology Information Service, Western New York Nuclear Research Center, Inc., Power Drive, Buffalo, N. Y. 14214. (FEM)

Phosphorus-33 Availability

The Isotopes Development Center at Oak Ridge National Laboratory (ORNL) has announced the availability of ^{33}P for purchase by approved commercial buyers. Phosphorus-33 is better for certain applications than ^{32}P because of its longer half-life (25 vs. 14.3 days) and lower maximum beta energy (0.25 vs. 1.7 Mev). Results of the use of experimental quantities of ^{33}P in research programs, together with literature references, indicate that this radioisotope will be useful in several fields, e.g., biology, medicine, agriculture, and ecology. The U. S. Atomic Energy Commission has approved the following prices:

- $3300 per batch for a two-cycle (6-week) reactor irradiation (~200 mc of separated ^{33}P).
- $3500 per batch for a three-cycle (9-week) reactor irradiation (~300 mc of separated ^{33}P).
- $35 per millicurie in less than batch quantities, the minimum order being $35.

For additional information, including complete isotope sale-price lists, contact the Isotopes Sales Department, Isotopes Development Center, Oak Ridge National Laboratory, P. O. Box X, Oak Ridge, Tenn. 37830.

Isotope Shipments in 1967

The U. S. Bureau of the Census reports that $15.7 million worth of radiation sources and other radioactive materials produced from purchased isotopes was shipped in the United States in 1967. This includes shipments to U. S. government agencies and exports of selected important atomic energy products manufactured in privately owned establishments.

Radioactivity Standards Available

A number of radioactivity standards are available from the Office of Standard Reference Materials, Room B308, Chemistry Building, National Bureau of Standards (NBS), Washington, D. C. 20234. Two recently announced standards are:

^{109}Cd–^{109m}Ag: NBS standard No. 4202 consists of $^{109}CdCl_2$ sandwiched between two layers of 0.006-cm-thick polyester tape. The standard had a gamma-ray emission of ~1×10^5 gammas/sec in December 1967. It costs $88 per unit.

^{226}Ra Solution: The radium gamma-ray solution standards NBS Nos. 4955 through 4963, which were originally made up in 1947, have been recalibrated using the more accurate working standards now available. The energy emission rates of the current working standards were determined by measurement with the national radium standards in the NBS radiation balance. Nominal ^{226}Ra contents of NBS Nos. 4955 and 4956 are 0.1 and 0.2 μg, respectively. For NBS Nos. 4957 through 4963, the nominal contents range from 0.5 μg for No. 4957 and 50 μg for No. 4963. The recalibrated ^{226}Ra standards cost $58 per unit, weigh approximately 5 g each, and are supplied in flame-sealed glass ampules.

ORNL Stable-Isotope Inventory Augmented

Recent additions to the ORNL enriched stable-isotope inventory include the following

Isotope	Purity, %	Amount available, g
^{34}S	33.5	28.0
^{34}S	44.6	24.9
^{100}Mo	97.4	80.7
^{102}Pd	56.6	0.07
^{145}Nd	80.6	1.7
^{204}Pb	70.9	114.8
^{204}Pb	99.7	2.3

(PSB)

^{125}I Source for Dental Radiography

A ^{125}I source that fits inside the mouth and produces a panoramic picture of the entire upper or lower jaw was demonstrated in Stockholm by Prof. C.-O. Henrikson of the Swedish School of Dentistry. (A similar source and X-ray pictures of the teeth made with it were reported by R. Söremark, Use of ^{125}I for Intraoral X Radiography, in *Proceedings of Symposium on Low-Energy X- and Gamma Sources and Application,* Held at Illinois Institute of Technology Research Institute, Chicago, October 20-21, 1964, USAEC Report ORNL-IIC-5, 1965.) The source is contained in an aluminum tube 2 cm long and 0.5 mm in diameter and requires exposures of 8 to 40 sec.

High Energy Processing Corp.

The High Energy Processing Corp. of New Bedford, Mass., is offering its irradiation services at a special introductory rate. The company owns a 7-Mev linear accelerator and has access to lower voltage equipment and a ^{60}Co facility. The introductory rate is $600 per month, for a minimum of 6 months, which includes up to 6 hr/month machine time and 10 hr/month consultation time. The standard price, which would go into effect at the end of the 6 months, is $100 to $150 per hour of machine time and $40 per hour of consultation time.

New Company Formed To Market Radiochemicals

The Amersham/Searle Corp., which began operation July 1, combines the UKAEA's Radiochemical Centre with Nuclear-Chicago Corp., the nuclear instrument subsidiary of G. D. Searle & Co. The new company is based in Chicago and will develop, make, and market radiation sources, radiochemicals, radiopharmaceuticals, and related products. John Kuranz,* senior vice president of Nuclear-Chicago, becomes president of Amersham/Searle; he is assisted by two Nuclear-Chicago executives (Robert Chudy as director of operations and Donald Zahorik as director of marketing) and one Amersham representative (Charles Evans as director of technical operations and manufacture). Sir Charles Cunningham, deputy chairman of the United Kingdom Atomic Energy Authority, is chairman of the board.

Initially Amersham/Searle will handle marketing for the Western Hemisphere. Any products developed, including labeled therapeutic and diagnostic agents at present not routinely marketed, will be made available to the Radiochemical Centre on an exclusive basis. Sales of the Radiochemical Centre in the United States have been near $1 million annually; Nuclear-Chicago has marketed about 65% of the products represented by this sum.

Gamma Process Company, Inc., Expansion

Gamma Process Company, Inc., is exploring acquisition and merger opportunities with a number of companies that can logically benefit from radiation processing, i.e., companies in the plastics, cosmetics, medical supplies, enzymes, and packaging fields. The company has the demonstrated technical radiation know-how to improve these products and broaden their markets and profitability. Gamma Process will continue to provide irradiation services for industry and research.

The first full-scale nuclear radiation processing facility in the United States is to be built by Gamma Process for the New York State Atomic and Space Development Authority. The $1.2 million plant, to be located at the Malta Test Station, in Saratoga County north of Schenectady, is expected to be in full operation in September 1969. Groundbreaking is scheduled to take place in October 1968. The initial capacity will be 500 thousand curies of ^{60}Co, to be increased later to up to 2 million curies. IsoNuclear Corporation, a wholly owned subsidiary, will operate the plant, which will process chemicals, plastics, medical equipment, disposable surgical equipment, cosmetics, and enzymes.

Union Carbide To Market "Minigenerator"

To help support the nucleonics offerings in high school and basic college science curriculums, Union Carbide's Sterling Forest Research Center is marketing a miniature version of a radioisotope generator which will permit demonstration of some simple radioisotope experiments with a minimum of laboratory equipment. The two systems being used are ^{137}Cs–^{137m}Ba and ^{113}Sn–^{113m}In, and the "Minigenerators" (Fig. 1) contain general-license amounts of activity.

The column of the generator is sized so that it can be used as a point source of radiation and can be placed directly in the counting chamber of a radiation detector. A plastic squeeze bottle, charged with the eluting solution, can be used to force the solution through the generator in about 30 sec. This permits easier handling of the eluant and faster elution of the generator.

A major advantage of the Minigenerator is that it is an inexpensive source of short-lived radioisotopes. It can deliver well over 1000 times its loading during its useful lifetime, which is many years for the ^{137}Cs–^{137m}Ba generator and about 1 year for the ^{113}Sn–^{113m}In generator. In addition, the generator can be eluted whenever desired and repeatedly during a given laboratory period.

*Mr. Kuranz, until recently, served as chairman of the USAEC's Advisory Committee on Isotopes and Radiation Development.

Fig. 1 Minigenerator.

^{137}Cs–^{137m}Ba

The 30-year ^{137}Cs–2.6-min ^{137m}Ba generator is attractive as an educational aid because of the contrast in half-lives. The long-lived parent provides a column that can be used for years, while the 2.6-min half-life of the daughter is ideal for decay and buildup studies. After being eluted, the generator returns to secular equilibrium within a few minutes; in fact, it can be milked every 2.6 min with a yield of 50% of the ^{137m}Ba activity produced when equilibrium is established. Hence it can be milked many times even during a very short laboratory period—a virtual faucet of a short-lived radioisotope!

Specifications

Parent nucleus	^{137}Cs (30 years)
Daughter nucleus	^{137m}Ba (2.6 min)
Type of emitted radiation	Gamma ray
Energy of radiation	662 kev
Eluant	0.04*M* HCl + 0.9% NaCl
Elution volume	2 to 5 ml
Specific activity of daughter	Carrier free
Activity of parent	0.9 μc

^{113}Sn–^{113m}In

The 115-day ^{113}Sn–100-min ^{113m}In generator complements the ^{137}Cs–^{137m}Ba generator for educational use in several ways. Indium-113*m* has a longer half-life (long enough to perform experiments that require an appreciable portion of a laboratory period), and it emits a gamma ray whose energy is slightly more than one-half that of ^{137m}Ba (ideal situation for instrumental calibration and gamma-ray–matter interaction studies).

Specifications

Parent nucleus	^{113}Sn (115 days)
Daughter nucleus	^{113m}In (100 min)
Type of emitted radiation	Gamma ray
Energy of radiation	393 kev
Eluant	0.04*M* HCl + 0.9% NaCl
Elution volume	2 to 5 ml
Specific activity of daughter	Carrier free
Activity of parent	9 μc

Table 1 lists typical experiments that can be performed with the Minigenerators. Detecting and

Table 1 Radioisotope Experiments

Experiment	Manipulation of generator	Purpose
	^{137}Cs–^{137}Ba	
1. Operating characteristics of a radiation detector	Point source	To determine the operating voltage of a radiation detector
2. Gaussian or normal distribution	Point source	To investigate the statistics of radioactive measurements
3. Generator elution	^{137m}Ba eluted from generator	To determine the amount of eluant required for maximum yield of ^{137m}Ba
4. Parent–daughter equilibrium	Source after elution	To study the secular equilibrium between parent and daughter
5. Half-life of ^{137m}Ba	^{137m}Ba eluted from generator	To determine the half-life of ^{137m}Ba
6. Absorption of gamma photons	Point source	To determine the half-value layer of lead for gamma rays
7. Energy of gamma rays	Point source	To measure gamma-ray energies using an oscilloscope

(Table continues on the next page.)

Table 1 (Continued)

Experiment	Manipulation of generator	Purpose
	$^{113}Sn-^{113}In$	
1. Operating characteristics of radiation detector	Point source	To determine the operating voltage of a radiation detector
2. Resolving time of radiation detector	^{113m}In eluted from generator	To determine the resolving time of a radiation detector by using the split-source method
3. Gaussian or normal distribution	Point source	To investigate the statistics of radioactive measurements
4. Generator elution	^{113m}In eluted from generator	To determine the amount of eluant required for maximum yield of ^{113m}In
5. Parent–daughter equilibrium	Source after elution	To study the secular equilibrium between parent and daughter
6. Half-life of ^{113m}In	^{113m}In eluted from generator	To determine the decay rate of ^{113m}In
7. Inverse-square law	Point source	To determine the effect of distance from a radioactive source on the intensity of its gamma radiation
8. Absorption of gamma photons	Point source	To determine the half-value layer of lead for gamma rays
9. Energy of gamma rays	Point source	To measure gamma-ray energies, using an oscilloscope
10. Compton scattering	Point source	To detect scattering of gamma rays passing through matter
11. Indium hydroxide precipitation	^{113m}In eluted from generator	To study the use of radioactive tracers in a chemical reaction
12. Reactions between ions in aqueous solution	^{113m}In eluted from generator	To determine end products in a theoretical equation
13. Gaging	Point source	Density gaging
14. Tracing	^{113m}In eluted from generator	To trace inaccessible systems
15. Flow rate of fluids	^{113m}In eluted from generator	To measure flow rate within a closed system
16. Volume of a container	^{113m}In eluted from generator	To measure the volume of an irregular container
17. Amount of fluid in a container	^{113m}In eluted from generator	To determine the amount of fluid in a container
18. Washing efficiency	^{113m}In eluted from generator	To determine the efficiency of detergents and soaps
19. Diffusion	^{113m}In eluted from generator	To determine the diffusion of indium ions through a membrane
20. Wear studies	^{113m}In eluted from generator	To detect evidence of wear from friction

counting equipment that can be used include a scaler, scintillation detector system, dual timer, and G–M counter—all of which can be purchased from commercial vendors for a total less than $1000; however, for many experiments a simple G–M counter is adequate. For certain experiments a cathode-ray oscilloscope is also needed. The generators themselves sell for $40 each and are available from W. H. Curtin Scientific Supply Co., Houston, Tex., and Welch Scientific Supply Co., Skokie, Ill. (PSB)

AEC Activities

Radioisotope Licensing-Procedure Revisions

The USAEC is considering establishing new types of broad licenses under which certain qualified licensees would have greater flexibility in their operations. Institutions with small- or intermediate-scale programs would benefit primarily.

At present, broad licenses authorized by Title 10 in Part 33, *Code of Federal Regulations,* are issued only to institutions that have a radiological safety officer and, in some cases, an isotope committee with sufficient technical skill and administrative authority to ensure that the radioisotopes are used safely and in compliance with AEC regulations. They allow institutions to possess multicurie quantities of radioisotopes but are useful only where there are large-scale programs that undergo frequent changes. Most radioisotope licenses now issued specify the kind of isotopes, chemical and physical forms, possession limits, uses, and users, and they must be amended when a licensee's program changes.

The proposed revisions will provide for Types A, B, and C broad licenses. The Type A license would be a combination and revision of the existing broad licenses, with Types B and C scaled-down versions of Type A. For Type A to be issued, a radiological safety officer and an isotope committee would be required in all cases. Types B and C would authorize users to possess 10,000 and 100 times, respectively, the quantities permitted to be inhaled by a person occupationally exposed for a year to the relevant concentration levels set forth in 10 CFR Part 20, Appendix B, Table I. More than one radioisotope would be allowed, but in proportionately reduced amounts. For a Type B license, the applicant must have a qualified radiological safety officer and administrative procedures to control uses and users. For a Type C license, each applicant must have a scientific background and training in radiation protection. Types B and C licenses would not relax safety standards or lessen licensing requirements in exchange for training, experience, facilities, and equipment.

The proposed revision of Part 33 would also expand the scope of the uses—i.e., research and development—authorized in all broad licenses. Under the revision, radioisotopes could also be used for purposes authorized by the Atomic Energy Act of 1954, including agricultural and industrial.

The Commission would consider applications to be for broad-scope licenses, even though they are not filed as such, if the application shows that the requirements for any such license have been satisfied. The scope of the licenses would normally cover the entire program of an applicant, but there would be exceptions for operations requiring specific licensing under other AEC regulations. These would include such uses as the manufacture and distribution of items to persons exempt or generally licensed, industrial radiography, human use, and operations such as tracer studies in the environment involving direct release of radioisotopes and the use of large sources in irradiators. It is expected that the proposed revision would improve the regulatory process by reducing administrative requirements on both the licensees and the Commission.

The proposed revision follows comments and recommendations by the Commission's Advisory Committee on Isotopes and Radiation Development and by the Commission's ad hoc Radioisotopes Licensing Review Panel.

Changes in Self-Luminous-Product Regulations

The AEC has amended its regulations to increase the amount of radioactive ^{147}Pm that can be used in aircraft self-luminous safety devices used without a specific license from 100 mc to 300 mc. These signs and markers, in which beta radiation from the promethium acts on phosphors to provide light, contribute to the safety of air travel since they continue to operate if a plane's electrical system fails. The amendment resulted from a petition from Minnesota Mining & Manufacturing Co. of St. Paul, Minn., who want to increase the initial brightness of the devices to meet requirements of the Federal Aviation Administration as well as to ensure a longer useful life of the signs and markers. The Commission has concluded that the increase in quantity of promethium will not endanger public health and safety.

The AEC is also proposing a new class exemption for self-luminous products containing tritium, ^{85}Kr, and ^{147}Pm which meet specific safety standards. In the past the Commission has considered each petition for exemption of a product containing such materials individually and amended its rules to accommodate each type of product. The change in procedure is expected to expedite the regulatory process and save

time for both licensees and the Commission without relaxing safety requirements. Excluded from the class exemption are toys and adornments; also, the Commission may deny an application for a specific license if the end uses of the product cannot be reasonably foreseen. Petitions for rule making to exempt the use of such materials in toys or adornments or in self-luminous products that do not meet the standards for the new class exemption would continue to be considered by the Commission on a case-by-case basis.

Under the new rules a manufacturer seeking a license to distribute self-luminous products containing these materials would be required to submit (1) results of tests which demonstrate that the exempt units would contain the radioactive material and radiation therefrom for the life of the product under both normal and the most severe conditions of handling, storage, use, and disposal; and (2) details on the intended use, design, and expected lifespan of the product; the amount and type of radioactive material; and the estimated radiation exposure to ensure that exempted products would not endanger public health. Petitions already received for exemptions for self-luminous products, e.g., sportsmen's compasses, transistor radio dials, automobile windshield-wiper and headlight switch knob markers, safety signs, and control panels, would be covered.

Proposal To Abolish Specific License for ^{125}I Users

Proposed changes in AEC regulations would eliminate the need for a specific license for physicians, clinical laboratories, or hospitals for the use of ^{125}I in tests outside the human body. Iodine-131 is already exempted from specific license requirements for in vitro use, and small quantities of either isotope can be used on human beings for certain medical purposes. Under the new regulations, both isotopes could be possessed and used in vitro in prepackaged units not exceeding 10 μc each with total amounts not exceeding 200 μc at any one time. The user would be required to register with the AEC before receiving or using the radioisotopes. Manufacturers and distributors of either product would still need a specific license.

^{252}Cf Source To Be Tested for Mineralogy Applications

The use of ^{252}Cf, a radionuclide that spontaneously fissions to produce neutrons, will be investigated for possible use in mineralogy. The source is to be made by the USAEC's Savannah River Laboratory in South Carolina. The U. S. Geological Survey will study the usefulness of the source in determining moisture content of rocks and in locating minerals. [See W. C. Reinig, Advantages and Applications of ^{252}Cf as a Neutron Source, *Nuclear Applications*, 5: 24-25 (1968); J. P. Nichols, Design Data for ^{252}Cf Neutron Source Experiments, *Nuclear Applications*, 5: 382-387 (1968) for discussion of ^{252}Cf sources.]

International Project on Fish Irradiation

The governments of Iceland and the United States are cooperating in a 1-year project with the Food and Agriculture Organization of the United Nations and the International Atomic Energy Agency to irradiate fish and extend its refrigerated shelf life. The irradiation unit, provided by the USAEC, is a 35-kilocurie ^{60}Co irradiator designed for installation aboard a mother ship of a trawler fleet. The unit arrived in Reykjavik on May 31. The government of Iceland provided installations, laboratories, manpower, space, and other services for the experimenters. However, Iceland has no plans to commercialize the process at present.

Two Icelandic fishing specialists came to the United States to study irradiation methods. The USAEC provided a cost-free expert from BNL to Iceland for the irradiator installation, and John Kaylor of the Bureau of Commercial Fisheries Technological Laboratory, Gloucester, Mass., spent 1 month in Iceland to oversee the initial period of the project.

(FEM)

Price Increase for ^{137}Cs

The AEC has proposed an increase in the price of ^{137}Cs, which is used in industrial radiography, nuclear gages, and medical therapy and which has potential applications in radiation processing of chemicals, medical supplies, and foods. The new and old price schedules are:

Proposed new		Old	
Order size, curies	Unit price, $/curie	Order size, curies	Unit price, $/curie
0–50,000	3.00	0–10,000	0.50
50,001–200,000	2.00	10,001–50,000	0.45
>200,000	0.85	50,001–200,000	0.35
		>200,000	0.125

In 1965 the AEC decreased prices on ^{137}Cs, which is recovered from nuclear fuel-processing wastes, to levels substantially below production and distribution costs. This was done to stimulate the market prior to construction and operation of a planned private-industry recovery plant, which was subsequently canceled. The new price schedule would recover full costs.

Radioisotopic Dating of Paintings

The USAEC and the National Gallery of Art are each contributing $25,000 for the first year of a 3-year project at the Mellon Institute in Pittsburgh, which will include the study of activation analysis for use in detecting art forgeries. Activation analysis of paints from different centuries has shown* differences in the trace-element concentrations. The objective of the project is to develop isotope dating techniques applicable to lead-bearing pigments, metals, or other art materials, and methods for identifying sources of pigments using activation analysis.

^{129}I Price Lowered

The USAEC has reduced the price on large amounts of ^{129}I to 50 cents per milligram for quantities of 10 g or more. The previous price was 75 cents per milligram for amounts >500 mg. The price reduction is possible because batch orders of 10 g or more can be produced more economically than smaller amounts. Further information is available from the Isotopes Sales Department, Oak Ridge National Laboratory, P. O. Box X, Oak Ridge, Tenn. 37830.

*F. Lux and L. Braunstein, Radioisotopic Dating of Paintings: Analysis of White Lead from the Paintings of Dutch and Venetian Masters of the 16th and 17th Centuries, *Angewandte Chemie, International Edition in English*, 5: 679 (1966) [*Isotopes and Radiation Technology*, 5(4): 288 (Summer 1968)]; J.P.W. Houtman and J. Turkstra, Neutron Activation Analysis and Its Possible Application for Age Determination of Paintings, in *Radiochemical Methods of Analysis*, Vol. I, Symposium Proceedings, Salzburg, 1964, pp. 85-103, International Atomic Energy Agency, Vienna, 1965 (STI/PUB/88).

Agreement with Idaho Approved

The USAEC has approved the proposal by the State of Idaho for an agreement under which Idaho will assume part of the AEC's regulatory authority over the use of radioactive materials in that state. The transfer of regulatory responsibility will include licensing, rule making, and enforcement in the uses of radioisotopes, the source materials uranium and thorium, and small quantities of fissionable materials. There are 42 AEC licenses in Idaho for the use of radioactive materials. The Department of Health is the agency responsible for administering the state's radiation control program, which will cover the use of radioactive materials as well as X-ray machines and other sources of radiation not regulated by AEC.

License Fee Schedule Adopted

The USAEC is adopting a schedule of fees to be charged for licenses to construct and to operate nuclear reactors and other production or utilization facilities; for by-product material (radioisotopes) of 100 thousand curies or more in sealed sources; for special nuclear material in quantities sufficient to form a critical mass (except Pu–Be neutron sources); and for waste-disposal licenses specifically authorizing the receipt of radioactive materials for commercial disposal. The fee schedule is contained in a new Part 170 of Commission regulations entitled "Fees for Facilities and Materials Licenses under the Atomic Energy Act of 1954, as Amended."

General

Radioisotope-Powered Watch Patented

A patent (U. S. No. 3,370,414) has been granted to Benrus Watch Co. for a radioisotope-powered watch. The isotope used is ^{99}Tc, which has a half-life of 500 thousand years. The emitted radioactivity is counted electronically, and the watch hands are moved by a small battery that powers the electronic circuits. The watchcase is said to be an effective biological shield, and the hazard to the wearer of such a wristwatch would be negligible. Although the amount needed for a watch would cost only about 1 cent, a

watch of this type would not at present compete with others on a cost basis. The first use of a radioisotope-powered timepiece would probably be at such locations as unattended weather stations or in space probes.

Modern Trends in Activation Analysis

The third conference on modern trends in activation analysis was held at the National Bureau of Standards Laboratory in Gaithersburg, Md., Oct. 7–11, 1968. Besides the NBS, the sponsors of the conference were the USAEC, the IAEA, and Eurisotop. Each day of the conference was devoted to a specific subject: nuclear reactions, applications, radiochemical separations, radiation detection systems, and computation and data handling.

New England Nuclear Corp. To Expand

New England Nuclear Corp. has bought out Picker's half interest in their joint venture, NEN–Picker Radiopharmaceuticals, and "Picker" will be dropped from the name. Production will be increased with the installation of a $425,000 cyclotron, planned for 1969, in a new $300,000 facility. The firm plans to continue buying enriched stable isotopes from the AEC.

New ^{90}Sr Therapy Source

Atlantic Research Corp. is working on a new ^{90}Sr therapy source that will have glass and steel walls <0.005 in. thick. The walls are strong enough for safe containment but thin enough to allow efficient use of the beta radiation, for example, in extracorporeal irradiation of the blood of leukemia patients. A portable (50-lb) unit containing a 5-curie source will be the first application of the new capsule. A 1-curie research unit, which the company plans to adapt for irradiation to suppress antibodies in organ transplants, will also be offered. Some 8000 hospitals are expected to be interested in the sources.

Canada Considers Radiation Processing of Food To Have Good Prospects

At AECL's Commercial Products Ltd., the food industry is considered to show the most potential for applications of large radiation sources. Canada has a short growing season, long shipping distances, and a significant food export business, and increasing the shelf life of foods is particularly important under these conditions.

X-Ray Fluorescence in Industry

A Panel on Radioisotope X-Ray Fluorescence Applications met in Vienna in May 1968. The Panel, organized by the IAEA to evaluate developments in the field and advise on the best ways to promote their adoption, is composed of authorities from Australia, Austria, Canada, France, Japan, Poland, the United Kingdom, and the United States. Dr. Henry Seligman, IAEA Deputy Director General for Research and Isotopes, in a talk before the Panel, recalled that it had taken industry more than a decade to pick up from the laboratory many ideas about radioisotope X-ray fluorescence applications. With the advice of the Panel, the IAEA hopes that such applications can be made available in developing countries to assist in industrialization.

Packaging Radioactive Materials

A new British Standard specifies methods that may be used to test the suitability of packaging for the transport of radioactive materials, including large radioactive sources:

BS 3895—Methods for the assessment of packaging for the transport of radioactive materials: Part 2 Fissile materials and large sources: (Metric units).

The standard takes into account International Atomic Energy Agency Regulations for the safe transport of radioactive materials (IAEA Safety Series No. 6, 1967 edition) and provides a basis for United Kingdom authorities to approve packaging designs.

Appendixes are provided to assist the designers of packaging, giving advice on the mechanical and thermal protection required for the packaging under accident conditions, guidance on calculative assessment, and other general information relating to the test requirements.

Copies of BS 3895: Part 2: 1968 may be obtained from the BSI Sales Office, 101/113, Pentonville Road, London, N1. Price 20 shillings plus postage.

American Nuclear Corp. To Build Irradiators

The American Nuclear Corp. of Oak Ridge, Tenn. (P. O. Box 426, Oak Ridge, Tenn. 37830), was selected to design and build irradiators for two of the country's leading research organizations—the Battelle Memorial Institute in Richland, Wash., and the Salk Institute for

Biological Studies in La Jolla, Calif. The Battelle unit was delivered in July. It is loaded with an annular source of ^{137}Cs and has both electrical and manual controls for complete fail-safe and automatic operation. The Salk Institute unit was delivered in September. It is loaded with 5000 curies of ^{60}Co, which will be brought into the irradiation position pneumatically. It is designed for use in animal studies and delivers a conical radiation beam. The company has also delivered four 120-curie ^{137}Cs calibration sources to the Navy.

Union Carbide Plans 250-Thousand-Curie Irradiator

Union Carbide Corporation has filed an application with the U. S. Atomic Energy Commission to operate a 250-thousand-curie ^{60}Co irradiator at its large petrochemical complex at Institute, W. Va. (near Charleston). The facility is expected to be ready for operation in the fall and will be used to treat part of the plastics that Carbide makes and markets for various commercial uses. The irradiator is believed to be the largest in the United States to be used in chemical production.

Every possible safety precaution has been designed into the irradiation system to prevent accidental exposure. The entire system will be enclosed in a four-story-high tower with a 5-ft-thick wall of reinforced concrete. When not in use the radioactive cobalt rods will be stored deep in the ground. A series of special locks will prevent anyone from entering the vault when the rods are above ground. The system will be operated by remote controls located in a service building adjoining the tower. All plans were designed in accordance with AEC regulations and in consultation with AEC personnel, and the AEC will check the facility before granting the license.

Isotopes in State Hospital

The first isotope facility at a state mental hospital was put into operation in April at the Elgin State Hospital, Ill. Brain scanning and blood-volume determinations are being performed. Equipment for various medical and surgical tests and treatment, including a multiprobe thyroid scanner and a spectroscaler, is available.

Nuclear Activation Center in New York State

The State (New York) Office of Atomic and Space Development has executed a $200,000 contract with New York University covering participation in construction of a $1 million Nuclear Activation Center at the university's campus in the Bronx. Construction will begin early in 1969 and is expected to be completed by late 1970.

The principal component of the center will be a 250-kw(e) TRIGA research reactor, for use in nuclear activation research and analysis in such fields as atmospheric pollutants, criminalistics, and medicine, e.g., trace-metal diseases. The reactor fuel is being provided without charge by the U. S. Atomic Energy Commission. The center will be available for education, research, and training to state agencies and to educational nonprofit institutions within the state. The center's facilities and services will be available to private enterprise and federal and other government agencies on a reasonable contract basis.

Radioisotope-Actuated Sounding Device

An acoustic device that is actuated by the energy emitted by a radioisotope is available from Sanders Nuclear Corp., Nashua, N. H. It has a nominal power output of 10 watts. Since its frequency is adjustable over the range 8 to 12 kc, several units can operate in a small area without interfering with each other. The unit is said to operate at depths ranging down to somewhat greater than a mile. The transducer has a 3-mile slant range with all-direction sound propagation above the horizon.

New Spanish Radiation Centers

Three new radiation centers are being proposed by the Junta de Energía Nuclear for Spain. One is expected to be in Barcelona for radiation sterilization of medical products. The other two, planned for Granada and Murcia, are for food processing. Foreign companies will be allowed to compete for the work only if the Junta lacks the required know-how.

(RHL and MG)

Recent Accessions of Isotopes Information Center

Progress in Atomic Medicine: Nuclear Medicine and Recent Advances, Volume 2. Edited by John H. Lawrence, M.D., Director, Donner Laboratory and Donner Pavilion, Lawrence Radiation Laboratory, Berkeley, Calif., Grune & Stratton, Inc., New York, 1968. (ix + 274 pp., $12.75)

This particular compilation, Vol. 2 of the series, comprises eight reviews by particularly competent authors and is well documented with references. The chapters are:

1. "Hormone Studies in Health and Disease Using Radioimmunological Assay," by F. C. Greenwood and J. Landon. The use of radioisotopes for these very sensitive assays has opened up whole new frontiers of investigation and diagnosis and has made possible the evaluation of the effect of therapy in various disorders. The basis of radioimmunoassay is described, and individual assays are discussed, e.g., insulin, glucagon, human-growth and plasma-growth hormones, ACTH, thyrotropin, parathyroid hormone, human placental lactogen, and human gonadotropins. 238 references.

2. "Studies of Pulmonary and Cardiac Function Using Short-Lived Isotopes Oxygen-15, Nitrogen-13, and Carbon-14," by J. B. West. The possibility of using these three relatively new isotopes is significant in that they represent the three principal gases of cardiorespiratory interest—oxygen, carbon dioxide, and nitrogen. They are all positron emitters and can therefore be detected by coincidence techniques. Their use potential has only been scratched, primarily because of the need for a medical cyclotron facility for their production. But this requirement is rapidly being satisfied. Excellent results are obtainable with ^{15}O-labeled CO_2 for measurement of blood flow and ventilation in the lung, thus permitting detection of heart disease, lung disease, and cardiac shunts. Oxygen-15 can also be used to follow the fate of inhaled oxygen, carbon monoxide, carbon dioxide, and water vapor in the body. Nitrogen-13 can be used to investigate lung function. Carbon-11 is particularly useful for measuring sizes of body pools of CO_2 and rates of exchange between them. Red-cell volume measurements involve ^{11}CO, and metabolic studies utilize ^{11}C-labeled organic compounds. 47 references.

3. "Progress in Determining Bone Marrow Distribution in Vivo," by D. C. Van Dyke, H. O. Anger, and Y. Yano. The use of the short-lived positron emitter ^{52}Fe and the scintillation camera to take "pictures" is showing considerable promise in the in vivo studies of the distribution of hemopoietic bone marrow. It permits the physician to understand the changes occurring in conditions affecting the bone marrow and helps in management of the individual patient. 25 references.

4. "Quantitation of Red Cell and Heme Production and Destruction Using Radioisotope Kinetics," by H. S. Winchell. The application of tracer isotopes kinetics to red-cell behavior has permitted quantitation of red-cell and heme production and destruction. The isotopes ^{32}P, ^{51}Cr, ^{14}C, ^{3}H, ^{53}Fe, ^{59}Fe, and ^{75}Se are used for studying problems of circulating-red-cell and cohort-red-cell populations, i.e., total numbers, turnover rates, and rates of production. 37 references.

5. "The Present Status of Therapy with Radioactive Isotopes," by L. R. Wasserman and J. L. Glass. This is an excellent summary, which starts with a review of the basic principles of therapeutic applications and then lists the various isotopes and source forms for both internal and external applications. Beta and gamma sources are included, as are contact, interstitial, and extracorporeal techniques. 318 references.

6. "Recent Advances in Scanning," by P. V. Harper, A. Gottschalk, and R. N. Beck. The objective of scanning—to collect the most information about the isotope distribution in the patient in the least time—has given rise to many different kinds of devices. Some of the requirements are reciprocal and vary from situation to situation with different pathological conditions, radiation emitters, pharmaceutical agents, and scanners. These various factors are discussed and related to each other. The meaning of the term "scanning" is also discussed and placed in proper perspective. 10 references.

7. "Kinetics of Metabolic Processes as Studied with Radioactive Tracers," by E. Manougian. This paper covers the mathematical theory of metabolic process kinetics, using compartmental models, and shows the value of radioisotope tracers (e.g., ^{51}Cr, ^{14}C, ^{45}Ca) in such studies. 54 references.

8. "Space Radiation and Other Medical Aspects of Space Travel," by C. A. Berry and S. B. Curtis. The volume concludes with an interesting look to the future in a discussion of space medicine. The effects of space radiation on the skin, central nervous system, eyes, ears, nose, throat, and blood and on the gastrointestinal, cardiovascular, genitourinary, and musculoskeletal systems are described. Also included is

a discussion of the radiation environment found in space. 26 references.

All in all, this is a very fascinating volume and should be of considerable interest and help to scientists concerned with the areas covered. (PSB)

Industrial Uses of Radioisotopes, Proceedings of Symposium conducted by Unisearch Limited at the University of New South Wales, Kensington, October 25–27, 1967, Southern Cross International, Sydney, 1968. (198 pp.; $5.95, Australian currency)

For the purpose of gathering evidence of some of the peaceful applications of atomic energy in Australia, a symposium on the industrial uses of radioisotopes was conducted by Unisearch Limited at the University of New South Wales in October 1967. Contributors consisted of senior scientists and industrialists from the various states of Australia, who presented, in lay terms, a survey of the many ways in which radioisotopes have been put to use in Australian industry. Seventeen papers are presented, starting with an overview of the general field, by J. N. Gregory, and ending with D. L. Ford's evaluation of the symposium. In between, there are discussions of food irradiation, radiation-sterilization of sutures, radiation-induced polymerization and grafting, radioisotopes in analysis (e.g., of ores and slurries by X-ray fluorescence; or of extent of mixing or flow patterns through the use of tracers), radioisotopes for radiography, radioisotopes in the paper industry, and radioisotopes in the steel industry.

Since these are papers from a symposium, there is some duplication of coverage, but this is not serious. Also, there are no references cited; however, many specific examples of applications, with organizations and personnel involved, are listed, and there are a few line drawings and a number of good photographs. For a quick survey of industrial applications, this is a handy booklet, particularly for the "man on the street."

(PSB)

Isotopes Information Center Reports*†

L. V. Metlitskii, V. N. Rogachev, and V. G. Krushchev, Radiation Processing of Food Products, USAEC Report ORNL-IIC-14, Machine Translation from the Russian made by the Computing Technology Center of Union Carbide Nuclear Division, edited by Martha Gerrard, F. E. McKinney, P. S. Baker, and Eugen Wierbicki (U. S. Army Natick Laboratories), Oak Ridge National Laboratory, 1968. (CFSTI, 115 pp.)

Isotopes Development Center Reports

R. E. Lewis and T. A. Butler, Reactor Production and Characterization of Cadmium-109, USAEC Report ORNL-4247, Oak Ridge National Laboratory, 1968. (CFSTI, 12 pp.)

E. E. Ketchen, Preparation and Forming of High-Purity Cerium Oxysulfide, USAEC Report ORNL-TM-2117, Oak Ridge National Laboratory, 1968. (CFSTI, 12 pp.)

L. O. Love and W. A. Bell, Review of ORNL Electromagnetic Separations Program, 1966–67, USAEC Report ORNL-4271, Oak Ridge National Laboratory, 1968. (CFSTI, 78 pp.)

Division of Isotopes Development Reports

L. E. Preuss, C. Artman, C. Bugenis, and H. Collins, A Compilation of Beta Excited X-Ray Spectra; Part II (^{90}Sr–^{90}Y, ^{63}Ni, ^{204}Tl), USAEC Report TID-22361(Pt. 2), Edsel B. Ford Institute for Medical Research, 1967. (CFSTI, 226 pp.)

L. E. Preuss, W. S. Toothacker, and C. K. Bugenis, Polychromatic and Monochromatic X-Ray Absorptiometry Using Beta Excited, Electron Capture, and Other Isotopic X-Ray Sources, Final Report, September 1, 1965–July 14, 1966, USAEC Report COO-1239-16, Edsel B. Ford Institute for Medical Research, 1968. (CFSTI, 61 pp.)

L. J. Ronsivalli, V. G. Ampola, F. J. King, and J. A. Holston, Study of Irradiated-Pasteurized Fishery Products, October 1, 1966–September 30, 1967, USAEC Report TID-24256, Bureau of Commercial Fisheries, Technological Laboratory, 1967. (CFSTI, 81 pp.)

B. Sellers, H. H. Wilson, and C. A. Ziegler, Program To Explore and Evaluate Potential Applications of Radioisotope Excited X-Ray Fluorescence Techniques in Criminalistics and National Security, USAEC Report ORO-3561-1, Panametrics, Inc., 1967. (CFSTI, 18 pp.)

U. S. Atomic Energy Commission, Radioisotopes, Production and Development of Large-Scale Uses, USAEC Report WASH-1095, 1968. (Superintendent

*All CFSTI reports are available for $3.00 (full size) and $0.65 (microfiche) from Clearinghouse for Federal Scientific and Technical Information, National Bureau of Standards, U. S. Department of Commerce, Springfield, Va. 22151.

†International Atomic Energy Agency (IAEA) publications may be obtained from the National Agency for International Publications, Inc., 317 East 34th Street, New York, N. Y. 10016.

of Documents, U. S. Government Printing Office, Washington, D. C. 20402, $0.50, 41 pp.)

U. S. Atomic Energy Commission, Seventh Annual AEC Food Irradiation Contractors Meeting, Washington, D. C., September 27–28, 1967, USAEC Report CONF-670945, 1967. (CFSTI, 243 pp.)

J. I. Wagner and J. S. Milau, Failure of Cobalt-60 Radiation Sources, USAEC Report BNL-50099, Brookhaven National Laboratory, 1968. (CFSTI, 39 pp.)

R. I. Weller, Development of Methods of Industrial Control Measurements of Low-Level Contamination of Materials, Final Report, USAEC Report NYO-3540-1, Franklin and Marshall College, 1967. (CFSTI, 31 pp.)

Conferences

Isotopes and Radiation in Entomology, Symposium Proceedings, Vienna, December 4–8, 1967, International Atomic Energy Agency, Vienna, 1968 (STI/PUB/166). ($9.00, 428 pp.)

Isotope and Radiation Techniques in Soil Physics and Irrigation Studies, Symposium Proceedings, Istanbul, June 12–16, 1967, International Atomic Energy Agency, Vienna, 1967 (STI/PUB/158). ($9.00, 446 pp.)

P. Polishuk (Ed.), Nucleonics in Aerospace, Proceedings of the Second International Symposium on Nucleonics in Aerospace, Columbus, Ohio, July 12–14, 1967, Wright-Patterson Air Force Base, Ohio, Plenum Press, New York, 1968. ($17.50, 373 pp.)

Miscellaneous

The following three documents were developed jointly by the Division of Vocational and Technical Education of the U. S. Office of Education and the Division of Nuclear Education and Training of the U. S. Atomic Energy Commission and are available from the Superintendent of Documents, U. S. Government Printing Office, Washington, D. C. 20402, for the prices shown. They are intended as the basis of an 80-hr training course designed to provide the basic knowledge and skills necessary for the beginning radiographer:

Industrial Radiography—Manual, Report OE-84036, 171 pp., $1.25

Industrial Radiography—Student Guide and Laboratory Exercises, Report OE-84035, 87 pp., $0.55

Industrial Radiography—Instructor's Guide, Report OE-84034, 54 pp., $0.40

L. A. Bray, Recovery of Cesium from Purex Alkaline Waste with a Synthetic Zeolite, USAEC Report BNWL-288, Battelle–Northwest, Pacific Northwest Laboratory, 1967. (CFSTI, 16 pp.)

G. Brunner, X-Ray Fluorescence Analysis with Radioisotopes (in German), *Isotopenpraxis*, 3: 165-174 (1967).

C. B. Clifford, Design and Fabrication of a Prototype Laminated Uranium Metal Shipping Cask for Large Shipments of Co-60, USAEC Report KY-521, Paducah Gaseous Diffusion Plant, 1967. (CFSTI, 38 pp.)

J. Dauphin, Polymerization of Ethylene by Gamma-Rays, III Kinetic Model (in French), *Int. J. Appl. Radiat. Isotop.*, 18: 773-777 (1967).

D. L. Davis and F. L. Oetting, Analysis of Americium by Calorimetry, USAEC Report RFP-895, Dow Chemical Co., Rocky Flats Division, 1967. (CFSTI, 8 pp.)

R. R. Edwards, Method of Tracing Iodine Using I-129, U. S. Patent No. 3,339,072, to Nuclear Science and Engineering Corp., Aug. 29, 1967 (filed June 14, 1962).

J. W. Franklin, Apparatus for Measuring the Quality of a Cigarette Manufacturing Process, U. S. Patent No. 3,336,478, to American Tobacco Co., Aug. 15, 1967 (filed Aug. 20, 1964).

R. E. Garrett and W. K. Talley, Use of Radioisotopes in Selecting "Crisphead" Lettuce for Harvesting, *Nucl. Appl.*, 3: 745-749 (1967).

S. Godar, Process for the Production of the Phosphorus Isotope ^{33}P, British Patent No. 1,057,154, to European Atomic Energy Community (Euratom), Feb. 1, 1967 (filed June 16, 1964).

Gulf General Atomic, Inc., Activation Analysis Publications Prepared by the Activation Analysis Staff of Gulf General Atomic Incorporated, Report GA-7989 (Rev. 2), 1968. (Available on request from Gulf General Atomic, Inc., P. O. Box 608, San Diego, Calif. 92112, 27 pp.)

J. Holzhey, E. Oertel, and F.-W. Wagner, A Device for Radiometric Measurements of the Thickness of Materials in Hot Rolling in the Interval from 1 to 10 mm (in German), *Isotopenpraxis*, 3: 33-34 (1967).

W. H. Johnston and F. W. Lampe, Production of Hydrazine, U. S. Patent No. 3,322,660, to Atomic Energy Commission, May 30, 1967 (filed Feb. 18, 1963).

O. A. Kuhl, Integrally Bonded Encapsulated Gamma Source, U. S. Patent No. 3,331,962, to Atomic Energy Commission, July 18, 1967 (filed Sept. 17, 1964).

J. W. McMillan, Radioactive Tracer Methods in Inorganic Trace Analysis: Recent Advances, Report AERE-R-5266, United Kingdom Atomic Energy Authority, Research Group, 1967. (HMSO,* 22 pp.)

National Canners Association, Investigations on the Removal of Radioactive Fallout from Vegetables and Fruits During Processing by Normal and by New or Modified Processing Operations, Final Progress Report, June 1, 1964–May 1, 1968, Report D-3017, 1968. (CFSTI, 82 pp.)

K. Nikolov, Z. Kolev, S. Mazhkarov, and K. Mutaftchiev, Method for Production of Carrier-Free F-18 from Neutron Irradiated Lithium Carbonate (in Russian), *Isotopenpraxis*, 2: 433-435 (1966).

*Her Majesty's Stationery Office, London.

Radiation Preservation of Food, Conference, Oak Ridge, Tenn., Feb. 2–3, 1967, USAEC Report CONF-670202, Southern Interstate Nuclear Board and Atomic Energy Commission, 1967. (CFSTI, 100 pp.)

C. R. Schafer, Measurement of the Mass of a Material in a Container by Radioactive Means, U. S. Patent No. 3,244,897, to Simmonds Precision Products, Inc., Apr. 5, 1966 (filed June 27, 1962).

L. B. Shappert and R. S. Burns, Indexed Bibliography on Transportation and Handling of Radioactive Materials, USAEC Report ORNL-NSIC-33, Oak Ridge National Laboratory and Battelle Memorial Institute, 1967. (CFSTI, 147 pp.)

L. Wasser and J. M. Prosser, Radioisotope-Powered Cardiac Pacemaker Program, Mid-Term Report, Phase I, USAEC Report NUMEC-3731-13, Nuclear Materials and Equipment Corporation, 1968. (CFSTI, 146 pp.)

P. M. Yavorsky, Efficient Beta-Ray Emission Source for Irradiation Applications, U. S. Patent No. 3,322,951, May 30, 1967 (filed Aug. 28, 1964).

RADIOACTIVE WEIGHING SYSTEM FOR MOVING VEHICLES

A method for weighing moving vehicles, especially railway freight cars, has been patented by J. J. Wright (U. S. Patent 3,132,357, May 5, 1964). The system employs a radiation source in the roadbed with the detector on an overhead bridge. The amount of radiation reaching the detector is directly related to the weight of the car and its load; when the dimensions and weight of the car are known, the weight of the load can be readily calculated. This is especially true since mass absorption coefficients of the usual load materials do not vary more than about 10% from their common average. According to Wright, railway freight cars moving at speeds up to 30 mph could be weighed. Investment is estimated at one-fourth of that of ordinary scales. Accuracy is sufficient to allow comparison with weights reported in waybills, to permit sidetracking of overweight cars, and to spot underloaded cars in order to increase their load and improve operation. (FEM)

ISOTOPIC CHROMATOGRAPHY (Chromatographie Isotopique). I. Elution: Quantitative Models; Transfer Equations

By C. Jousselin and C. Massot, Société Nationale des Petroles d'Aquitaine, Pau, 1968
134 pp. (in French; English translation, ORNL-tr-1950, by Martha Gerrard)

The National Aquitania Petroleum Company is active in the investigation of chromatography as a method for separating isotopes. The theoretical bases of the process are discussed in detail in this book. Two earlier reports from this company on chromatographic separation of carbon isotopes (SNPA-59, ORNL-tr-1435) and of neon and argon isotopes (SNPA-60, ORNL-tr-1631) were abstracted earlier in *Isotopes and Radiation Technology* [4(3): 258 (Spring 1967)]. (MG)

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1 **Progress in Medical Radioisotope Scanning (TID-7673),**
Oak Ridge Institute of Nuclear Studies, 1962

2 **Reactor Kinetics and Control (TID-7662),**
The University of Arizona, 1963

3 **Dynamic Clinical Studies with Radioisotopes (TID-7678),**
Oak Ridge Institute of Nuclear Studies, 1963

4 **Noise Analysis in Nuclear Systems (TID-7679),**
University of Florida, 1963

5 **Radioactive Fallout from Nuclear Weapons Tests (CONF-765),**
U. S. Atomic Energy Commission, 1964

6 **Radioactive Pharmaceuticals (CONF-651111),**
Oak Ridge Institute of Nuclear Studies, 1965

7 **Neutron Dynamics and Control (CONF-650413),**
The University of Arizona, 1965

8 **Luminescence Dosimetry (CONF-650637),**
Stanford University, 1965

9 **Neutron Noise, Waves, and Pulse Propagation (CONF-660206),**
University of Florida, 1966

10 **Use of Computers in Analysis of Experimental Data and the Control of Nuclear Facilities (CONF-660527),**
Argonne National Laboratory, 1966

11 **Compartments, Pools, and Spaces in Medical Physiology (CONF-661010),**
Oak Ridge Associated Universities, 1967

12 **Thorium Fuel Cycle (CONF-660524),**
Oak Ridge National Laboratory, 1968

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Oak Ridge Associated Universities, 1968

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